Assessment of Ozonation and Biofiltration as a Membrane Pre-treatment at a Full-scale Drinking Water Treatment Plant

by

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AUTHOR'S DECLARATION

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

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Abstract

Membrane technologies are gaining popularity for drinking water treatment; however, fouling remains a major constraint as it can increase operational cost and shorten membrane service life. An important source of foulants for low pressure membranes (LPMs) is natural organic matter (NOM) which is present to varying degrees in all surface waters. Membrane fouling attributable to NOM can be managed by using appropriate pre-treatment(s). Among the new developments in membrane technologies for drinking water applications has been the integration of different pre-treatment processes in order to achieve optimal membrane performance and minimum lifecycle cost. The process combination of ozonation and biological filtration (biofiltration) appears to be a promising integrated pre-treatment for LPMs as both processes have been shown to individually be able to reduce LPM fouling. However, the process combination is neither commonly employed nor well-studied.

The goals of this research were to assess the fouling control capacity of ozonation-biofiltration as an integrated pre-treatment process for ultrafiltration (UF) membranes, evaluate the role of ozone in the ozonation-biofiltration-membrane (OBM) process combination, and investigate the effect of water quality and NOM on the process. The approach involved the operation of three UF pilot plants and long-term water quality and biomass monitoring at the Lakeview Water Treatment Plant (WTP), which is located in Southern Ontario and is one of the few WTPs in the world that employs an ozonation, biofiltration, and ultrafiltration process sequence. A novel Liquid Chromatography-Organic Carbon Detection (LC-OCD) method was used to characterize different NOM fractions, including biopolymers, humic substances, building blocks, low molecular weight (LMW) acids and humics, and LMW neutrals.

During this 16-month investigation, the ozonation-biofiltration process combination achieved good turbidity reduction but only minimal dissolved organic carbon (DOC) removal. In addition, the operation of ozonation (on vs. off) clearly impacted both biomass quantity and activity within the BACCs as measured by adenosine triphosphate (ATP) and fluorescein diacetate (FDA), respectively. This is because ozone can decrease the hydrophobicity of DOC in water as seen by a 43% reduction in specific ultraviolet absorbance through ozonation. Among all NOM factions measured by LC-OCD, biopolymers, which made up 13% of DOC, appeared to be the only one responsible for UF membrane fouling. An average of 60% of the biopolymers reaching the full- and pilot-scale UF membranes were retained. The concentration of biopolymers in membrane influent was found to be correlated to the hydraulically reversible fouling rate, while hydraulically irreversible fouling was largely affected by particulate/colloid content. The integrated ozonation–biofiltration pre-treatment process substantially reduced hydraulically irreversible fouling by removing substances measured as turbidity. Furthermore, ozonation was found to be able to enhance UF membrane fouling control as it can decrease biopolymer retention by downstream membranes (independently of biofilter efficiency).

This research provides valuable information for the water treatment sector on LPM fouling and its control. Overall, the full-scale integrated ozonation-biofiltration pre-treatment process successfully reduced downstream LPM hydraulically reversible and irreversible fouling, and as such the example of the Lakeview WTP can be used to guide designers of other municipal drinking water membrane installations. Information on the concentration and variation of biopolymers in source water is important for membrane water treatment applications, and biofilters should be optimized for better biopolymer removal. These findings provide useful insight into the design and operation of membrane water treatment facilities.

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List of Acronyms

AMWD: Apparent molecular weight distribution

AOC: Assimilable organic carbon

AOP: Advanced oxidation process

ATP: Adenosine triphosphate

BAC: Biological activated carbon

BACC: Biologically active carbon contactor

BB: Building blocks

BDOC: Biodegradable dissolved organic carbon

BOM: Biodegradable organic matter

BP: Biopolymers

DL: Detection limit

DOC: Dissolved organic carbon

DOM: Dissolved organic matter

EBCT: Empty bed contact time

EfOM: Effluent organic matter

EPS: Extracellular polymeric substances

FA: Fulvic acid

FDA: Fluorescein diacetate

GAC: Granular activated carbon

HPC: Heterotrophic plate count

HPM: High pressure membrane

HP-SEC: High performance size exclusion chromatography

HS: Humic substances

LC-OCD: Liquid chromatography - organic carbon detection

LMH: Liter per membrane area per hour

LMW: Low molecular weight

LPM: Low pressure membraane

MF: Microfiltration

MWCO: Molecular weight cut off
NF: Nanofiltration
NOM: Natural organic matter
OBM: Ozonation-biofiltration-membrane
OCD: Organic carbon detector
OND: Organic nitrogen detector
PVDF: Polyvinyl fluoride
RO: Reverse osmosis
SEM: Scanning electron microscope
SMP: Soluble microbial product
SUVA: Specific ultraviolet absorbance
TMP: Trans-membrane pressure
TOC: Total organic carbon
UF: Ultrafiltration
UMFI$_T$: Total unified membrane fouling index
UV: Ultraviolet
UVD: Ultraviolet detector
UV254: Absorbance of ultraviolet (UV) light at a wavelength of 254 nm
WTP: Water treatment plant
ΔTMP: Change in TMP during a filtration cycle

Chapter 1 Introduction

1.1 Problem Statement

Membrane technology for water treatment applications has developed dramatically in the last two decades. An important development in recent years has been the integration of pre-treatment strategies to improve membrane performance. Proper selection and operation of pre-treatment step(s) can enhance contaminant removal, reduce membrane fouling, extend membrane service life, and save on overall operating costs.

Intensive research on low pressure membranes (LMPs) is focused on the identification, quantification and control of foulants, especially organic foulants. Utilizing advanced natural organic matter (NOM) quantification tools such as Liquid Chromatography-Organic Carbon Detection (LC-OCD) (Huber *et al.*, 2011), some recent studies have demonstrated that biopolymers (e.g. polysaccharides, proteins) play an important role in both hydraulically reversible and irreversible fouling of LMPs (Hallé *et al.*, 2009; Peldszus *et al.*, 2011). Biological filtration has been identified as a promising pre-treatment option for LPMs because it can not only remove particles, but also reduce biopolymers as well as other organic matter and disinfection by-product precursors (Huck and Sozański, 2008).

Ozonation is a process reasonably commonly employed in water production. Its functions include disinfection, oxidation of micro-contaminants, enhancing biodegradability of organic matter and colour, and taste and odour control. It is common practice to follow ozonation with biological filtration in order to control biological stability in distribution systems (von Gunten, 2003; van der Helm *et al.*, 2009).

Ozonation-biofiltration as an integrated process for low pressure membrane pre-treatment appears quite promising as this pre-treatment technology has recently been demonstrated to successfully reduce membrane fouling and the combined process is able to produce high quality water, provide robustness, and produce water which consistently meets strict regulations. However, this integrated technology is neither commonly employed at full-scale membrane water treatment plants nor very well studied. Therefore, it is of great value to assess the fouling reduction capacity of this integrated pre-treatment process at full-scale. In addition, in order to better understand the mechanisms involved in fouling control, an investigation into the impact on NOM composition by the pre-treatment processes is required.

1.2 Research Motivation

The Lakeview Water Treatment Plant (WTP) in Mississauga, Ontario, Canada is one of the world's largest ultrafiltration membrane and biofiltration plants (Farr & Stampone, 2007). The advanced treatment train in this plant was commissioned in 2007, and it includes pre-chlorination, de-chlorination, ozonation, de-ozonation, biologically active carbon filtration, ultrafiltration (UF), and chlorination. This facility provides a rare opportunity to conduct research in assessing the effectiveness of ozonation-biofiltration as an LPM pre-treatment using natural water on a large scale.

It is of common interest for membrane technology researchers and practitioners to understand how and to what degree pre-treatment processes are able reduce membrane fouling for a particular type of water. The optimization of design and operation of such pre-treatment and membrane processes is also attracting substantial attention. In the case of the Lakeview Water Treatment Plant, the utility is constantly challenged to optimize its operation in order to improve energy efficiency and plant service life while maintaining the production of high quality drinking water. The municipality is also interested in knowing what processes can be changed or better designed in the plant’s future upgrade and expansion. The overall goal of this research was to provide useful insight into the operation and design of membrane water treatment facilities.

1.3 Approach

This research was conducted in the period from January 2013 to May 2014 at the Lakeview Water Treatment Plant (WTP) in Mississauga, Ontario, Canada. At the time of this research, this WTP had two process configurations. This study examined only the advanced treatment train described above.

Three UF membrane pilot-scale plants were operated, two with full-scale biofilter effluent and one with plant raw water, to achieve comparison in membrane performance and permeate quality. The UF membrane performance was assessed by monitoring flux, trans-membrane pressure (TMP) and permeability. LC-OCD analysis was used to measure the concentrations of different NOM fractions, including biopolymers, humic substances, building blocks, low molecular weight (LMW) acids and humics, and LMW neutrals. Some general water quality parameters such as total organic carbon (TOC), dissolved organic carbon (DOC), alkalinity, UV_{254} and specific ultraviolet absorbance (SUVA) were also measured. In addition, the quantity of viable cells present in biomass and activity of the cells were monitored using the surrogate compounds adenosine triphosphate (ATP) and fluorescein diacetate (FDA), respectively.

1.4 Research Objectives

In order to assess the fouling reduction capacity of full-scale ozonation and biological filtration pre-treatment processes for ultrafiltration (UF) membranes and understand how these pre-treatment processes control fouling, the following objectives were identified:

- Compare hydraulically reversible and irreversible membrane fouling without any pre-treatment to the ozonation-biofiltration combination as a pre-treatment.
- Investigate the effect of ozonation on both biofilter performance and membrane fouling.
- Monitor the biomass within the downstream biologically active carbon contactors (BACCs) with and without ozone being previously applied.
- Investigate the impact of natural organic matter (NOM) and water quality on membrane fouling and the pre-treatment processes.

1.5 Thesis Structure

Chapter 2 is an overview of published information related to this study. It introduces state-of-the-art low pressure membranes and biological filtration applications in water treatment, discusses the mechanisms and examples of using biofiltration as a low pressure membrane pre-treatment, and explains some of the analytical methods used in this study. Chapter 3 provides information on the Lakeview WTP and discusses the processes employed in the advanced treatment train. Each subsequent chapter was written in the form of a journal article, and each includes a dedicated methods section as well as results and conclusions (i.e. a paper-format thesis). Chapter 4 discusses the functions of ozonation and its role in the entire treatment sequence; particularly, how ozonation changes NOM properties and affects biomass and membrane fouling. In Chapter 5, the fouling reduction capacity by the integrated ozonation-biofiltration pre-treatment process is assessed. This chapter also reveals major foulants and their relationship with membrane performance. A final summary of conclusions and important recommendations are made in Chapter 6. The references from all chapters are compiled in a single list at the end of the thesis. Several appendices are provided for additional detail.

Chapter 2 Literature Review

2.1 Membrane Filtration in Drinking Water

2.1.1 Overview

Membrane technology emerged in the 1950s and its application in drinking water treatment began in the 1980s (Wiesner & Chellam, 1999). Since then the application of membrane processes in water treatment is becoming more prevalent, in particular microfiltration (MF) and ultrafiltration (UF). New regulations have triggered a greater interest in the application and development of membranes for researchers and practitioners. The advantages of employing membrane processes in water treatment are becoming more and more favourable for a sustainable future. Some of the advantages include better particle and pathogen removal, less chemical addition, smaller footprint and higher reliability. In addition, as the usage of membrane processes increases, the costs of the systems, both initial and long-term, have decreased dramatically (Freeman *et al.*, 2006).

2.1.2 Membrane Classification

Membranes are classified based on material of fabrication, pore size, surface characteristics, and operating conditions (Crittenden *et al.*, 2012). Based on material, commonly used membranes are divided into polymeric and ceramic groups. A more common way of membrane classification is based on pore size and operating pressure. With the pore size ranging from greater than 1 μm to less than 1 nm, pressure driven membranes are classified into four types: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) membranes (Figure 2.1). Among these four types of membranes, MF and UF membranes have operating pressure less than 0.5 MPa and therefore are referred to as low pressure membranes (LPMs), whereas the operating pressure of NF and RO membranes are greater than 500 kPa (0.5 MPa) and thus are classified as high pressure membranes (HPMs). Figure 2.1 also shows the different materials that these four types of membranes are able to reject. The selection of membranes is largely dependent on the rejection ability for each membrane type based on the application. For example, UF membranes are frequently employed to remove particles and pathogens from surface water and RO membranes are typically used in seawater desalination.

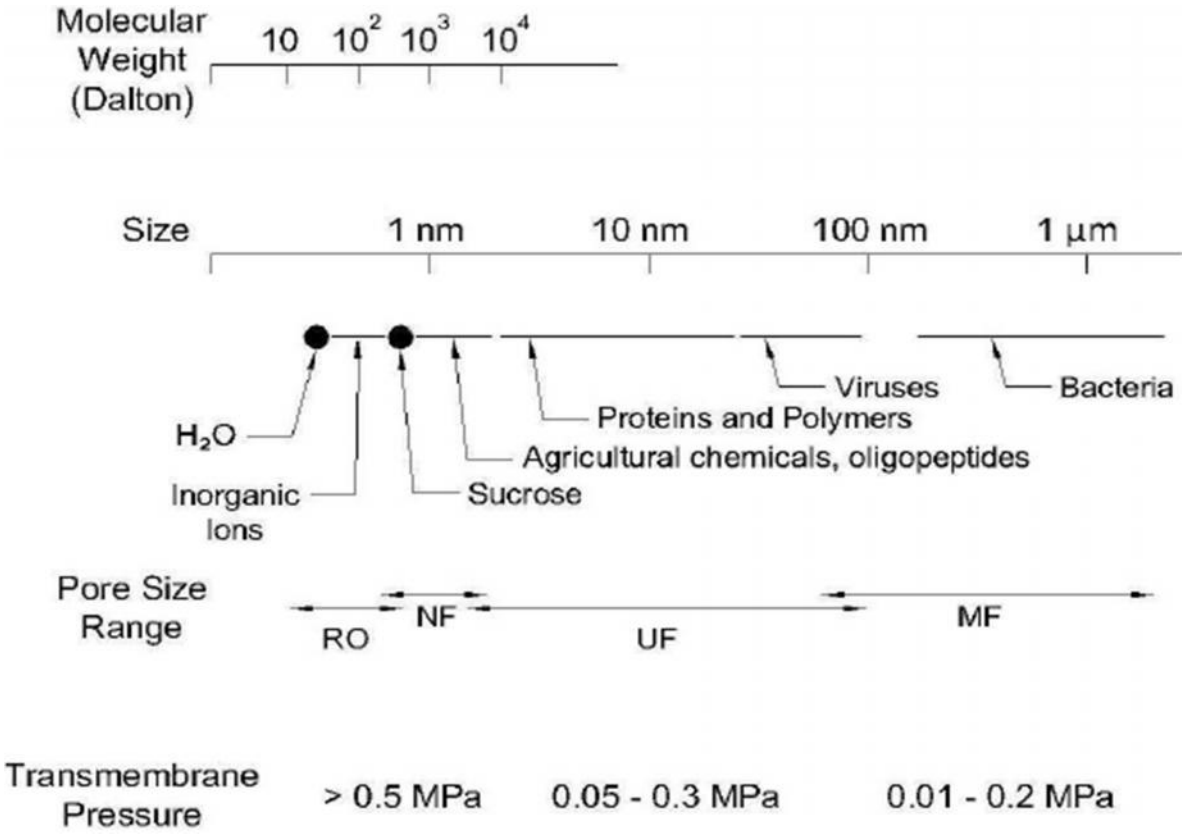

Figure 2.1 Classification of membranes based on pore size and trans-membrane pressure (Reprinted from El-Hadidy, 2011 with permission)

2.1.3 Membrane Rejection Mechanisms

The fundamental principal of membrane technology in drinking water treatment is to reject undesired particles, such as pathogens, allowing water and other dissolved matter to pass through. Membranes reject particles and suspended material based on the following mechanisms (van Der Bruggen *et al.*, 2002):

- Sieving/size exclusion is the most important membrane rejection mechanism. Particles larger than membrane pores will be retained on membrane surface and particles smaller than membrane pores will pass through. However, this phenomenon does not occur in all cases. Under some circumstances, flexible particles larger than membrane pore size are able to pass through membranes driven by pressure. Similarly, particles smaller than membrane pores can be retained as well due to interaction between particles and membrane surfaces.
- Hydrophobic adsorption is one type of such interaction. Certain types of particles can be adsorbed onto a membrane surface due to hydrophobic interactions. This rejection mechanism is important in the early stages of membrane operation; however, as the adsorption capacity is being reached, this mechanism becomes less effective in longer-term operation.
- Membranes are also able to reject particles by electrostatic interaction. As membrane surfaces are typically negatively charged, negatively charged particles can be rejected by electrostatic repulsion. Clearly, this mechanism is highly dependent on surface charge of both membrane and particles. Also, pH of the solution is an important factor for this mechanism.

2.1.4 Membrane Fouling

Typical membrane systems operate at constant pressure or constant flux. Flux is defined as the rate of water flow through membranes per unit area (AWWA, 2005). Membrane fouling is defined by AWWA (2005) as a gradual reduction in filtrate water flow rate at constant pressure or an increase in trans-membrane pressure at constant flux. It is the deterioration of membrane performance due to deposition of material on membrane surface and within pores. Fouling can develop through complete, intermediate and standard pore blocking and cake layer formation (Figure 2.2). Fouling remains to be a major challenge for the further adoption of membrane processes in water treatment since it leads to increases in operational complexity and maintenance costs, deterioration in productivity and shorter membrane service life.

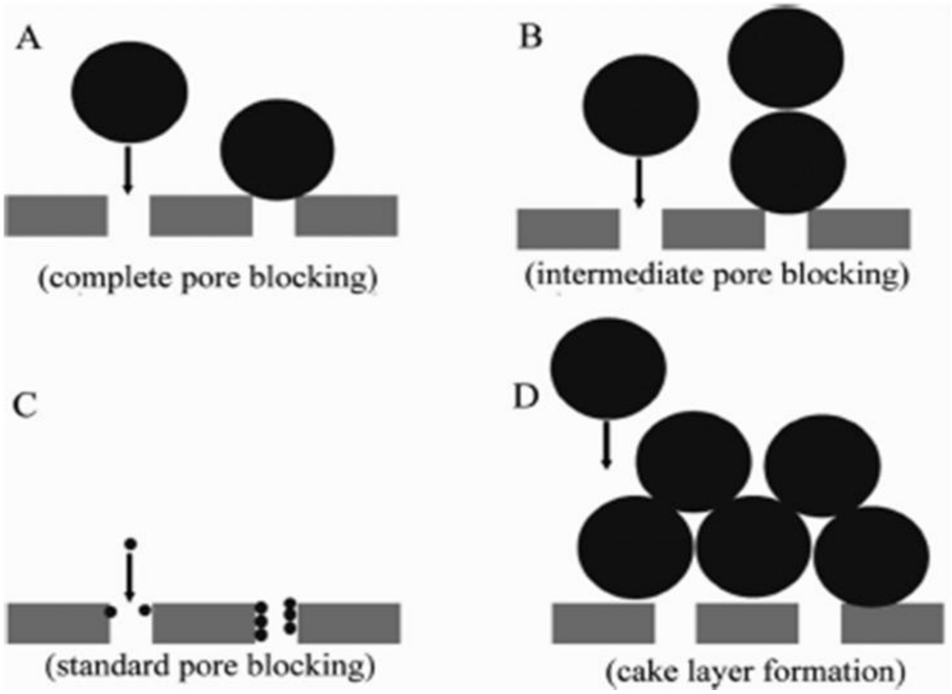

Figure 2.2 Schematic diagrams of four modes of membrane fouling. Reprinted with permission from Huang *et al.*, 2008. Copyright (2008) American Chemical Society

Based on reversibility, membrane fouling can be classified into hydraulically reversible and irreversible fouling in relation to backwash, and chemically reversible and irreversible fouling in relation to chemical cleaning (Figure 2.3). There are four classes of fouling based on foulant type (Mosqueda-Jimenez & Huck, 2006a; Gao *et al.*, 2011). Organic fouling is induced by natural organic matter (NOM) and/or effluent organic matter (EfOM); an important group of organic foulants are the biopolymers (Amy, 2008; Cai & Benjamin, 2011; Jiang *et al.*, 2010; Lee *et al.*, 2006). Inorganic fouling is also referred as scaling. It occurs when salt concentration exceeds its solubility limit. Inorganic fouling is common for HPMs due to their ability to reject ions and less common for LPMs as concentrations of rejected ions will not be high enough to cause severe salt precipitation (Her *et al.*, 2007). Colloidal fouling describes the deposition of inorganic colloids on the membrane surface to eventually form a cake layer. Colloidal fouling is typically easily removed by backwashing due to the weak chemical bonds (Howe & Clark, 2002). Biofouling is caused by adsorption of microbial cells onto the membrane surface via different types of intermolecular forces. It is less common in LPMs due to the use of chlorine in membrane cleaning (Baker & Dudley, 1998).

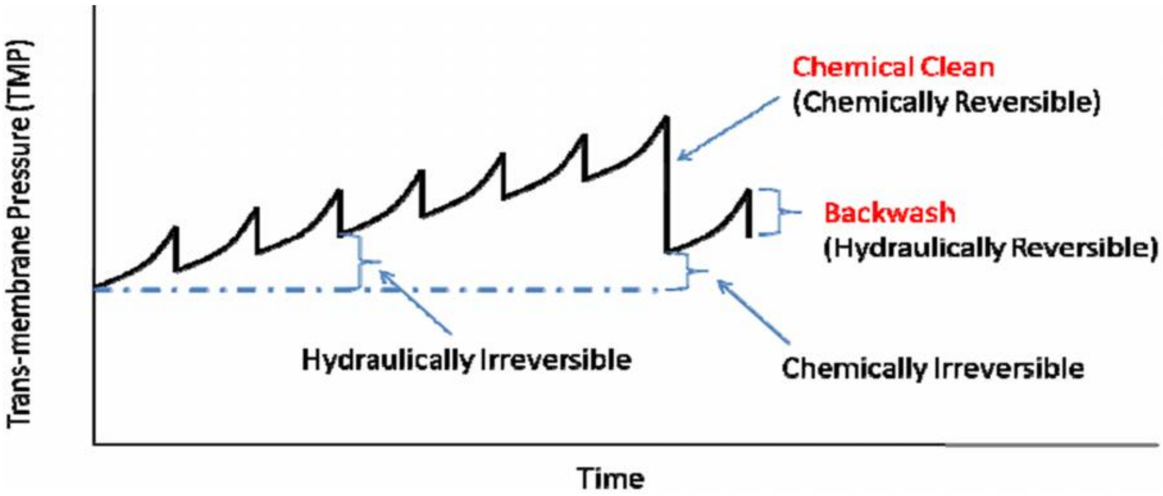

Figure 2.3 Classification of membrane fouling based on reversibility (Reprinted from Croft 2012 with permission)

2.1.5 Fouling Mitigation

As mentioned above and illustrated in Figure 2.3, membrane fouling can be controlled by backwashing and chemical cleaning. Backwashing involves reversing the permeate flow through membranes, and it is an essential component of operation for most membrane systems. By reversing the flow, some of the material deposited on membrane surface and within pores can be detached and flushed away. For typical water treatment membrane applications, backwashing is usually initiated every 30 to 60 minutes for a period of 2 to 3 minutes (AWWA, 2005). Some outside-in mode membrane systems also employ aeration during backwash for better removal of deposits from membranes. When backwash can no longer sufficiently remove foulant materials from membrane surface and pores, a chemical cleaning is required to partially or fully restore trans-membrane flux or pressure. A variety of agents can be employed for the chemical cleaning of membranes, including detergents, acids, bases, oxidizing agents, sequestering agents, and enzymes. Chlorine is commonly used for polysulfone (PS) or PS-derivative materials and polyvinylidene fluoride (PVDF) membranes (Zondervan & Roffel, 2007). Chemical cleaning is initiated when trans-membrane pressure exceeds 70 to 200 kPa (0.7 to 2 bar), depending on the particular membrane (AWWA, 2005).

Other than backwashing and chemical cleaning, membrane fouling can also be mitigated by pre-treating the feed water. Pre-treatment is a proactive measure for membrane fouling control. Proper pre-treatment can remove or alter significant portion of foulants before they reach the membranes, hence reducing fouling. In principle, pre-treatment of feed water can impact membrane filtration in three ways: altering particle size distributions, changing the interaction between contaminants and membrane surface, and suppressing undesirable microbial growth or removing biodegradable contaminants (Huang *et al.*, 2009). Membrane pre-treatment processes can not only reduce fouling but also enhance contaminant removal, and some processes may be able to achieve additional goals. The following processes may be used as pre-treatments for LPMs in water treatment:

- Adsorption pre-treatment refers to the use of absorbents, for example power activated carbon (PAC), prior to LPMs. PAC is the most commonly used absorbent, and it is used to remove trace contaminants, disinfection by-product (DBP) precursors and taste and odor causing compounds; these compounds are typically smaller than LPM pores, hence practically not removed by LPMs (Zhang *et al.,* 2003). Overall, the application of absorbents may or may not reduce membrane fouling.
- Pre-oxidation can also be used prior to LPMs as a pre-treatment. Some of the oxidants that can be used for this purpose include chlorine, permanganate, and ozone. Ozone seems to be a promising pre-treatment option for fouling control as it is able to partially oxidize NOM and alter the size distribution of dissolved organic matter (DOM). Several studies have demonstrated that ozone can reduce organic fouling in LPMs for water and wastewater treatment applications (Hashino *et al.,* 2001; You *et al.,* 2007; Kim *et al.,* 2008). The application of ozone as an LPM pre-treatment is further discussed in the next section.
- Coagulation, either applied "in-line" or associated with flocculation/sedimentation, can be an effective LPM pre-treatment. Finding the optimal coagulant dose is essential in this pre-treatment application because overdosing of a coagulant may adversely impact the downstream LPM fouling (Kimura *et al*., 2005; Howe *et* al., 2006; Yan *et al.*, 2008). In addition, the cost-effective coagulant dose for optimal LPM performance is often different from conventional coagulation practice for turbidity removal by downstream

filters. After reviewing several state-of-the-art pre-treatment options for LPMs, Huang et al concluded that coagulation is the most successful for fouling control (Huang *et al.*, 2009).

- The feed water can also be pre-filtered before it reaches LPMs. Pre-filtration can be in the form of granular media, a looser membrane or innovative material. Though pre-filtration by membranes with coarse pore matrices can significantly reduce downstream LPM fouling, it is frequently not cost-effective. Innovative pre-filtration material requires more research before being ready for large-scale industrial application. Granular media filtration can be effective for colloidal fouling control due to its ability to remove particles both larger and smaller than the membrane pore size (O'Melia, 1980; Sakol *et al.*, 2004), which if not removed will cause fouling. Recent studies (Park *et al.*, 2002; Hallé *et al.*, 2009; Huck *et al.*, 2011; Peldszus *et al.*, 2012; Rahman *et al.*, 2014) have identified biological filtration as a promising LPM pre-treatment since it can not only remove colloids like conventional granular media filters, but also remove biodegradable components of NOM, and hence control organic fouling. This innovative application is further discussed in later section.

In recent years, intensive research in membrane water treatment has been focused on the integration of different pre-treatment processes for better fouling control. Some examples for integrated LPM pre-treatment are: adsorption-flocculation (Vigneswaran *et al.*, 2004; Guo *et al.*, 2004; Shon *et al.*, 2005), ozonation-GAC filtration (van der Hoek *et al.*, 2000; Osterhus *et al.*, 2007; Treguer *et al.*, 2010; Qi *et al.*, 2011) and ion exchange-coagulation (Jarvis *et al.*, 2008; Huang *et al.*, 2012). In general, if membrane fouling can be effectively controlled by the integration of multiple pre-treatments, the lifecycle costs of membrane systems may be reduced in spite of the higher capital costs.

2.2 Ozonation in Water Treatment

2.2.1 Ozone Chemistry

Ozone is a highly reactive gas formed by electrical discharge in the presence of oxygen. Once enters in solution, ozone either undergoes direct oxidation or decomposes into hydraulic radicals (•OH). Disinfection typically involves molecular ozone, whereas oxidation processes frequently involve both molecular ozone and OH radicals (Langlais *et al*., 1991). Molecular ozone is a very selective oxidant; OH radicals react very quickly with many dissolved compounds in the water matrix (Hoigné, 1998). Other than disinfection, ozone can also oxidize a vast number of inorganic, organic and trace contaminants in water, such as metals, nitrite, ammonia, pesticides, solvents, taste and odor causing compounds and pharmaceuticals (von Gunten, 2003). In addition, ozone can also be applied in advanced oxidation processes (AOPs), which are referred to as processes that involve the formation of OH radicals as an oxidant. Ozone-based AOPs can oxidize contaminants that are resistant to ozone, such as chlorinated solvents and aromatic compounds (Karimi *et al*., 1997; Hoigné, 1998). Ozone also reacts with natural organic matter (NOM). Studies have demonstrated that ozone is able to change the composition of NOM (van der Helm *et al*., 2009; Treguer *et al*., 2010). In particular, ozone attacks double bonds and aromatic structures and breaks down large organic molecules, resulting in a decrease in hydrophobicity and the formation of by-products, such as hydroxyl, carbonyl and carboxyl groups (Urfer *et al*., 1997). Ozonation by-products are typically only of health concern when bromide is present in water (von Gunten, 2003; Hammes *et al*., 2006).

2.2.2 The Application of Ozone in Water Treatment

The first industrial ozone application was introduced in 1906; however, due to high capital costs and energy consumption by ozone generators, the application of ozone did not become widely accepted in North America until the awareness of chlorination disinfection by-products in the 1970s (Crittenden *et al.*, 2012). Since then, ozone was selected as a preferred alternative disinfectant for various water and wastewater treatment applications. In addition, as described above, ozone is also able to oxidize a variety of contaminants. However, one drawback of applying ozonation in water treatment is that it promotes biofilm development and microbial regrowth in distribution systems. This is because ozone breaks down large organic molecules, thus increasing the concentration of biodegradable organic matter (BOM) (van der Helm *et al.*, 2009). Therefore, it is common practice in water treatment to follow ozonation with biological filtration to control biostability in distribution systems by reducing the concentration of BOM through biological degradation. Biofiltration is also able to remove some of the organic ozonation by-products (Melin & Odegaard, 2000).

2.2.3 The Application of Ozone in Low Pressure Membrane Systems

As mentioned in the membrane section, ozonation seems to be a promising pre-treatment option for low pressure membranes in terms of fouling control due to its ability to alter the composition of NOM and the size distribution of DOM. Though not commonly employed in industrial applications yet, a number of studies have demonstrated the possibilities and benefits of using ozonation in low pressure membranes for different water treatment and reuse applications. For example, Hashino et al (2000 & 2001) used ozone-resistant MF membranes following ozonation for drinking water production, and found that the permeate flux was three to four times higher with pre-ozonation than without it. The authors also concluded that the reaction of ozone with DOM has to occur on the surface of the membrane to have such effect. In an investigation on an ozone-UF hybrid system, the researchers attributed flux enhancement to change in particle size distribution and membrane zeta potential (Hyung *et al*., 2000). Recent studies on water reuse applications used more advanced analytical methods. You et al (2007) tested ozonation prior to UF membranes to treat tertiary effluent from a wastewater treatment plant with bench-scale experiments, and found that with pre-ozonation, flux was improved by up to 60%. Scanning electron microscope (SEM) images on membrane surface also confirmed the finding by visually demonstrating less deposition on the surface of the membrane with pre-ozonation. Wang et al (2007) conducted similar experiments on secondary effluent. In addition, the authors also examined the apparent molecular weight distribution (AMWD) in water, and concluded that AMWD in water as well as membrane resistance were different with different ozone contact times. Since many traditional polymeric membranes are prone to damage by ozone, more attention is being focused on ceramic membranes for this type of ozone application (Karnik *et al*., 2005a; Kim *et al*., 2008). Similar findings were made in these studies.

2.3 Biological Filtration in Water Treatment

2.3.1 Overview

Derived from drinking water production filters, biological filtration effectively integrates physical and biological purification processes by making use of immersed filtration media on which microbial populations grow. Biofiltration has a history of over 50 years. During recent decades, biological rapid filtration processes have been integrated into practice (Urfer *et al*., 1997). Biofiltration can achieve a number of treatment goals. The primary goal is to reduce concentrations of easily biodegradable organic carbon in water, thus controlling significant microbiological growth within distribution systems (Rittmann, 1995). In addition to biodegradable organic carbon, other electron donors in water, such as ammonium, nitrite, ferrous iron, manganese and sulfides can also be removed by biofiltration (Rittmann & McCarty, 2001). In addition, biofiltration is applied in some new areas including the potential removal of trace contaminants such as pharmaceuticals, the removal of odorous substances such as geosmin, and the application as a membrane pre-treatment (Huck & Sozański, 2007).

2.3.2 Biological Processes in Biofilters

Biological processes in biofilters are complex, typically including attachment, detachment, biodegradation, growth, and decay (Figure 2.4). Microorganisms attach to media using extracellular polymer (EPS), which provides habitat and protection (Rittmann, 1995). The acclimation period of a biofilter may last several months until sufficient biofilm is established (Seredyńska-Sobecka *et al*., 2006; Velten *et al*., 2011). Microorganisms will grow on media surfaces when disinfectant is absent. Heterotrophic bacteria utilize organic carbon as an electron donor to grow and achieve biodegradation. Other nutrients, such as nitrogen and phosphorous are also critically important for the development of biomass (Brown, 2007). Typical surrogates for carbon sources available to microorganisms are assimilable organic carbon (AOC) and biodegradable dissolved organic carbon (BDOC). AOC is a measure of biodegradable material utilized by specific microorganisms which can be converted to cell mass (reported as a carbon concentration), while BDOC measures the organic carbon which is removed by heterotrophic microorganisms either under batch incubation conditions, or in specialized media columns (Rittmann, 1995). Ozonation prior to biofiltration enhances biological processes in biofilters because it increases the concentration of BOM in biofilter influent as measured by AOC and BDOC.

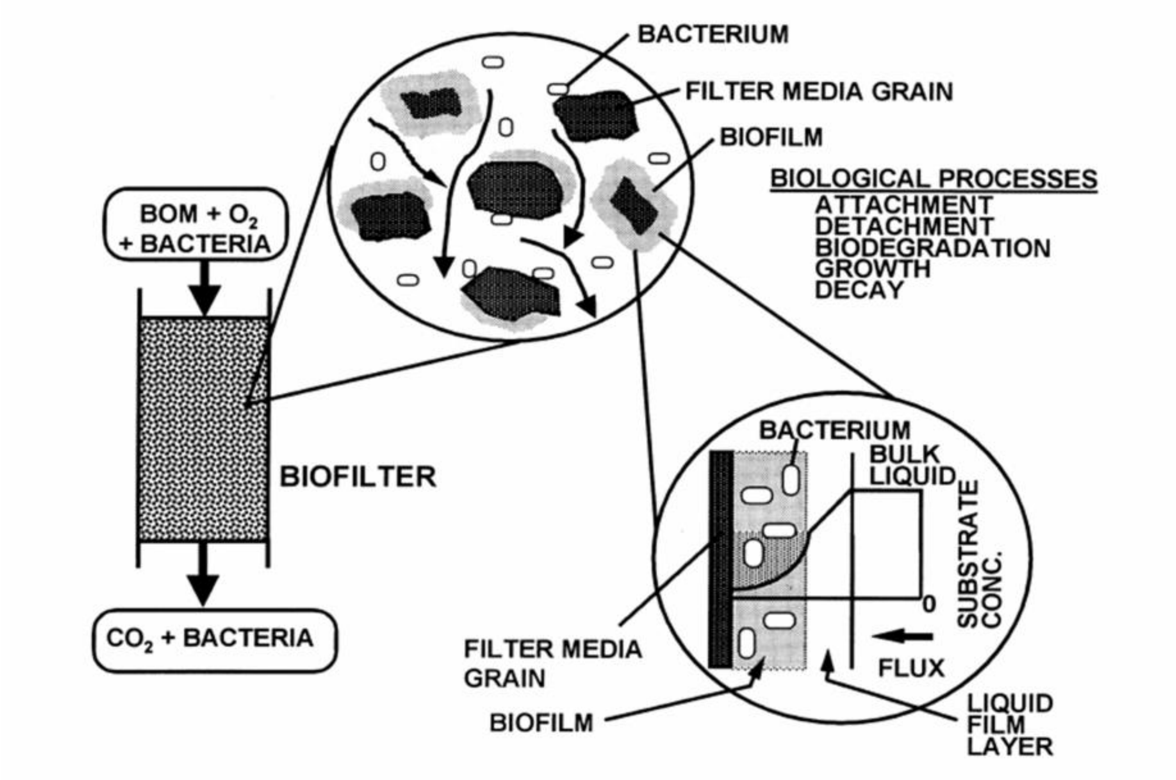

Figure 2.4 Biological processes in a biofilter. Reprinted from Hozalski *et al.,* 2001 with permission from *Elsevier*.

As the popularity of biofiltration has increased during recent decades, a number of methods have been developed or adapted to characterize the biomass attached to biofilter media. The total biofilm amount can be represented by a number of physical and physico-chemical parameters, such as biofilm thickness, total dry weight, total organic carbon (TOC) and chemical oxygen demand (COD) (Simpson, 2008). For example, the thickness of biofilm determines its mass transfer properties and is correlated with microbial growth (Lazarova & Manem, 1995); TOC represents approximately 50% of cell biomass (Harris & Kell, 1985). However, total biofilm amount tends to overestimate because not all biomass inhabitants are active. Viable biomass activity can be quantified by biochemical tests targeting on specific enzymes or products associated with microbial metabolism (Lazarova & Manem, 1995). Certain proteins and lipids are indicative of microbial activity and correlate well with substrate uptake rate (Wang *et al*., 1995). One example is adenosine triphosphate (ATP), which is an easy and quick method frequently used to provide an indication of viable biomass within biofilters (Pharand *et al*., 2014). ATP is the primary energy carrier for all living cells, and its concentration correlates well with the number of viable cells and oxygen uptake rate in biofilters (Magic-Knezev & van der Kooij 2004; Velten *et al*., 2011). Another example is fluorescein diacetate (FDA), which is a method adapted from soil samples to measure overall microbial activity. FDA can be hydrolyzed by important enzymes in microbial metabolism. Fluorescein is produced during the hydrolysis and can be measured by spectrophotometry (Adam & Duncan, 2001; Green *et al*., 2006). Biomass respiration potential (BRP) is another method for biomass activity quantification. It is sensitive and a good indicator of BOM removal (Urfer & Huck, 2001).

2.3.3 Operational Parameters Impacting Biofiltration

Operational parameters have a significant impact on the performance of biofilters including particle and BOM removal. Filter media is an essential element of a biofilter. Media type including particle size, surface area, texture, adsorption capacity, cost as well as media configuration are all important factors to consider when designing a biologically active filter. Despite the higher cost, granular activated carbon (GAC) biofilters have been demonstrated to perform better than sand-anthracite biofilters in terms of organics removal, adsorption of contaminants and biomass development (LeChevallier *et al*., 1992; Wang *et al.*, 1995; Urfer *et al*., 1997). Empty bed contact time (EBCT) is defined as the occupied volume of the filter media divided by the volumetric feed flow rate, and it is also an essential parameter for biofilter operation (Hozalski *et al*., 1995). Longer EBCT can improve organics removal

(LeChevallier *et al.*, 1992); however, once the optimal EBCT is reached, increasing EBCT can no longer improve biofilter performance (Zhang & Huck, 1996).

Similarly to traditional filters, biologically active filters also require periodic backwash to prevent accumulation of solids and headloss build-up. There are different strategies for backwashing, such as using air scour or not and using chlorinated or non-chlorinated water. A study compared different backwashing strategies on full-scale biofilters and concluded that BOM removal is insensitive to backwashing strategies (Emelko *et al.*, 2006). Using chlorinated water to backwash biofilters will largely destroy the biomass, especially on top of the filter; however, it does not have a significant impact on BOM removal (Miltner *et al.*, 1995). Temperature is highly important for microbial kinetics and therefore is very likely to have an impact on biofilter performance (Urfer *et al.*, 1997). Emelko et al (2006) also monitored biofilter performance in both warm (21-24°C) and cold (1-3°C) conditions, and observed lower oxalate removal and better GAC biofilter performance over sand-anthracite in cold conditions. Generally speaking, it is important to optimize these parameters if possible during design and operation of biofilters to achieve the best performance.

2.3.4 Biofiltration as a Low Pressure Membrane Pre-treatment

As mentioned previously, biofiltration can also be employed as a low pressure membrane pre-treatment for fouling control. This novel approach can also enhance contaminant removal, reduce disinfection by-products formation potential, and does not require chemical addition. Such benefits also allow biofiltration to be used as a pre-treatment for high pressure membranes in applications such as water reuse and desalination (Hu *et al.*, 2005; Chinu *et al.*, 2009; Mosqueda-Jimenez & Huck, 2009). Focusing on low pressure membranes and drinking water applications, a few studies have demonstrated the fouling control capacity by biofiltration and investigated its mechanisms. Park et al (2002) used biofiltration followed by MF membranes to treat surface water. The researchers observed less fouling in the biofiltration-MF hybrid system than MF alone, and attributed this effect to 55.9% turbidity and significant metal ion (Fe, Mn and Al) removal by the biofilter. Osterhus et al (2007) investigated particle removal by the process combination of ozonation-biofiltration-membrane (OBM), which utilized submerged polymeric UF membranes to treat natural organic matter (NOM) containing surface water. Fouling rates at different fluxes and different pHs were compared. The authors observed lower irreversible and higher reversible fouling rate at higher pH, which is believed to be caused by different oxidation products by ozonation and consequently change in size distribution and hydrophobicity. However, the authors neither further proved the hypothesis nor compared the fouling rate to the same UF system without pre-treatment. Hallé et al (2009) further investigated the fouling reduction mechanism by biofiltration using hollow fibre polymeric UF membranes. It was found that certain DOC fractions are more important than total DOC concentration for organic fouling. Using liquid chromatography-organic carbon detection (LC-OCD), the authors were able to identify biopolymers (i.e. polysaccharides and proteins) as the most important fraction of DOC that is responsible for organic fouling. In this study, biofiltration reduced downstream UF membrane fouling by removing over 90% of the turbidity and 40-60% of the biopolymers, which are largely biodegradable. Figure 2.5 shows the significant reversible and irreversible fouling reduction by biofiltration pre-treatment as indicated by trans-membrane pressure (TMP) data. In addition, the biofilter with a longer EBCT was found to have better fouling reduction capacity. The findings were confirmed by similar pilot-scale studies (Huck *et al.*, 2011;

Peldszus *et al*., 2012). Overall, biofiltration is a “green” and robust process as a low pressure membrane pre-treatment. More research is needed at relatively large scale to optimize the operation of biofilters for better fouling control and in its combination with other processes, such as ozonation.

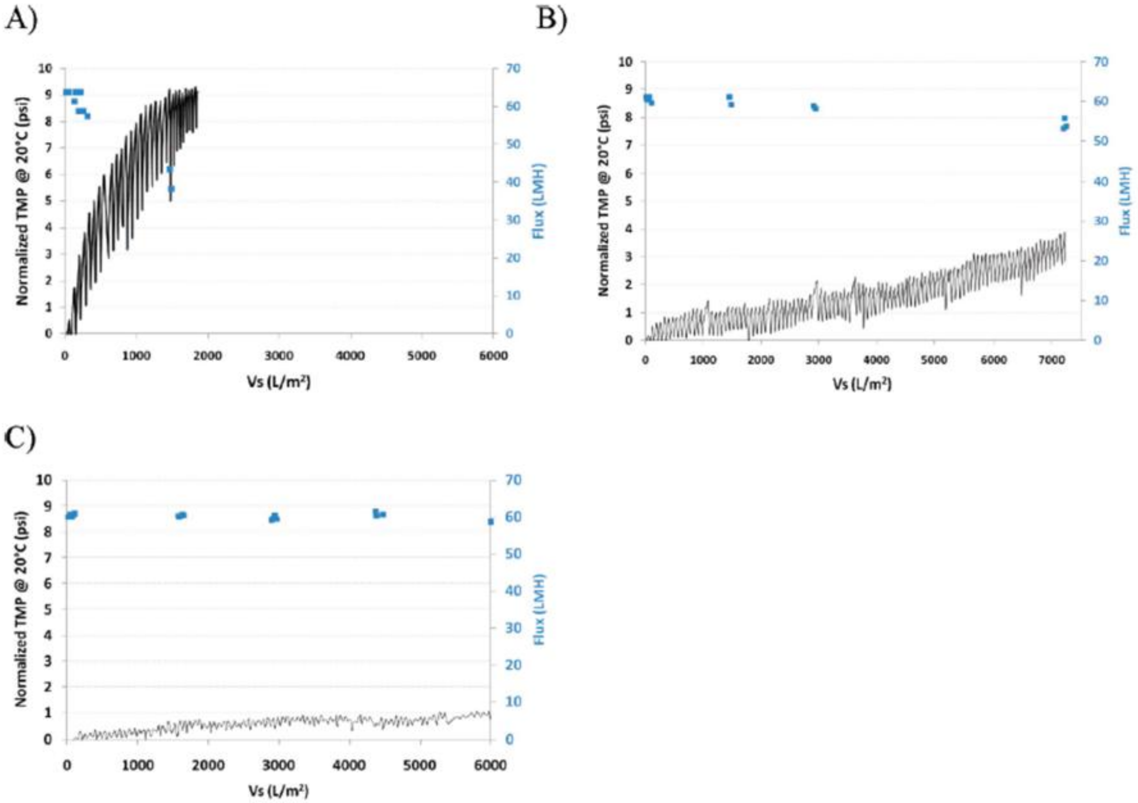

Figure 2.5 Effect of biofiltration pre-treatment on TMP of a UF membrane. The UF membrane was fed with the effluent from (A) a roughing filter, (B) biofilter 1 with EBCT 5 minutes and (C) biofilter 2 with EBCT 14 minutes. Reprinted with permission from Hallé *et al*., 2009. Copyright (2009) American Chemical Society

2.4 Natural Organic Matter and its Characterization

2.4.1 Introduction to Natural Organic Matter

Natural organic matter (NOM) is a complex heterogeneous mixture of naturally occurring organic material found in all water bodies. The sources of NOM can be allochthonous (soil derived decaying plant material), autochthonous (microbial by-products produced in-situ) and anthropogenic (wastewater discharge etc.). NOM can contribute to odour, colour, and taste problems in water and produce disinfection by-products when reacting with commonly employed drinking water disinfectants. It can also increase coagulant demand, affect corrosion and stability of water, and cause membrane fouling (Juhna & Melin, 2006). NOM composition is highly variable and complex. There are many forms of NOM classification; a simple and commonly used way is to divide NOM into six major compound classes including humic substances (humic and fulvic acids), hydrophilic acids, carboxylic acids, amino acids, carbohydrates, and hydrocarbons (Thurman, 1985).

2.4.2 NOM Characterization by Traditional Surrogate Methods

Due to many adverse effects of NOM in water treatment processes and its highly complex composition, it is important to characterize NOM in order to take proper measures to remove specific fractions. Some general parameters commonly used in water quality analyses can provide information on NOM. Total organic carbon (TOC) and dissolved organic carbon (DOC) are typical surrogates for total NOM quantity; they are measurements of CO_2 produced by the more or less complete oxidation of NOM. Ultraviolet (UV) absorbance at 254 nm (UV_{254}) is a water quality test parameter which utilizes UV light at the 254 nm wavelength to be able to detect organic matter in water. In particular, it is indicative of aromatic structures in NOM (Crittenden *et al.*, 2012). Specific UV absorbance (SUVA) is defined as UV absorbance at 254 nm divided by the DOC concentration of the water sample; it normalizes the bias of UV_{254} towards aromatic organic matter (USEPA, 2009). An SUVA value greater than 4 indicates the presence of highly hydrophobic and high molecular weight aquatic humics, while an SUVA value less than 2 is indicative of low hydrophobicity and low molecular weight (less humic content) (Edzwald & Tobiason, 1999). Smaller SUVA values suggest that the water sample contains more biodegradable organics (Juhna & Melin, 2006).

2.4.3 High Performance Size Exclusion Chromatography (HP-SEC)

Although traditional water quality parameters can provide some information on the nature and properties of NOM, one cannot speculate more on the composition of NOM and concentrations of different fractions with these general parameters. Therefore, a number of advanced analytical methods have been developed to better characterize NOM. High performance size exclusion chromatography (HP-SEC) is a powerful and relatively recent tool to characterize NOM by separating NOM into different fractions and measuring the concentration of each fraction. This process involves a mobile phase which passes through a stationary phase where molecules are separated according to their apparent molecular weight (AMW), shape, and chemical interaction. Large molecules tend to elute first, while smaller molecules elute later due to greater diffusion into pores of the column resin. Elution is then measured by means of an online detector, such as UV-vis and DOC detectors, with the strength of a signal in proportion to the concentration of a specific NOM fraction (Lankes *et al*., 2009). One specific example is the liquid chromatography-organic carbon detection (LC-OCD), which includes an organic carbon detector (OCD), an ultraviolet detector (UVD) and an organic nitrogen detector (OND). LC-OCD is a recently developed HP-SEC method and is gaining great popularity in water and wastewater applications due to its high sensitivity, multiple detectors and simple sample preparation procedures (Huber *et al*., 2011). LC-OCD can separate NOM into the following five fractions (Figure 2.6):

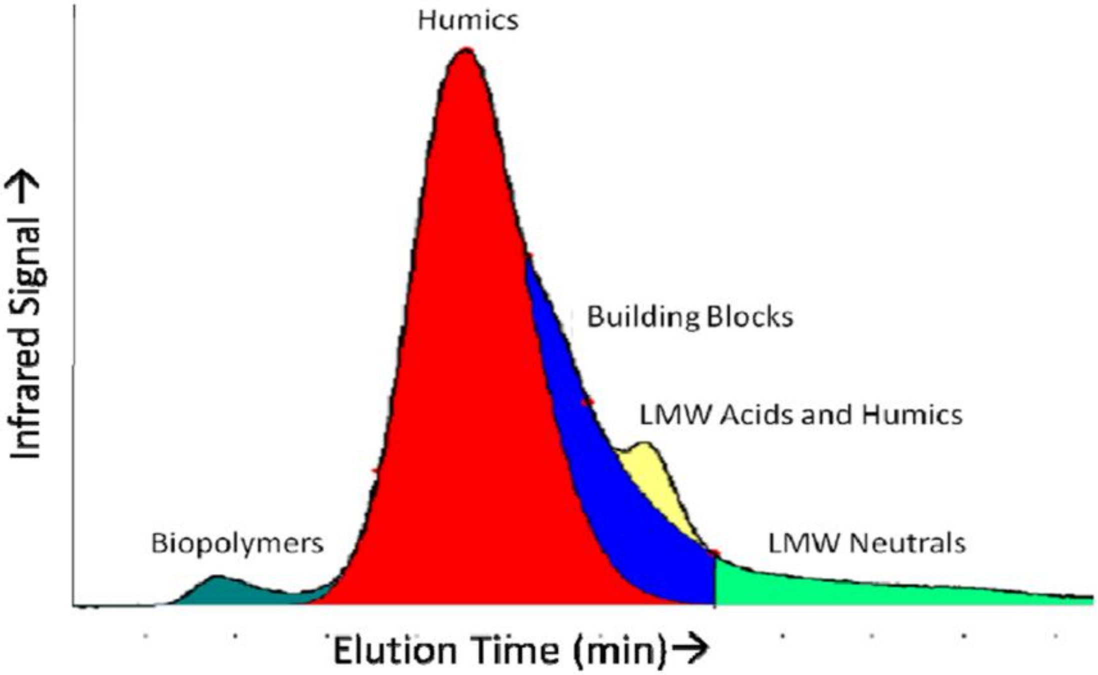

Figure 2.6 LC-OCD chromatogram OCD signal showing five different NOM fractions.
(Reprinted with permission from DOC-Labor Huber, 2010)

- Biopolymers: the fraction with the highest molecular weight (MW) (≥10,000 Da), and therefore are the first fraction to elute from the column. This fraction, which consists of polysaccharide and protein-like material, is highly hydrophilic. Due to its low hydrophobicity, the biopolymer fraction does not have a UV response (Huber *et al.*, 2011). Biopolymers are of biological origin, such as plants, animals and microorganisms, and hence can undergo biodegradation (Haberkamp *et al.*, 2011).
- Humic substances: the second fraction to elute and typically represent the largest peak of an LC-OCD chromatogram since humic substances are the dominant fraction of NOM for most natural waters. Humic substances include humic and fulvic acids; the average molecular weight of this fraction is approximately 1000 Da (Velten *et al.*,

2011). This fraction has a significant signal response to UV, which is attributable to its aromatic and unsaturated structures (Huber *et al*., 2011).

- Building blocks: the third fraction, which elute as a shoulder to the humic peak. Building blocks consist of degradation products of humic substances with a molecular weight range of 300-450 Da (Huber *et al*., 2011).
- Low molecular weight (LMW) acids: an aliphatic fraction which co-elutes with LMW humics as a compressed peak.
- LMW neutrals: the last fraction to elute which consist of alcohols, aldehydes, ketones, amino acids and sugars with no or low UV response (Huber *et al*., 2011).

Chapter 3 Background: The Lakeview Water Treatment Plant Process Configurations

3.1 Overview

This study was conducted at the Lakeview Water Treatment Plant (WTP), which is located on the north shore of Lake Ontario in Mississauga, Ontario (Figure 3.1). The WTP serves parts of Region of Peel and also provides water to York Region. At the time of this study, the Lakeview WTP had a capacity of 800 ML/d and consisted of a conventional treatment train and an advanced treatment train. The two treatment trains shared the 800 ML/d capacity equally. The conventional treatment train consists of the following processes: coagulation, flocculation, sedimentation, filtration with granular activated carbon (GAC), and chlorine disinfection. The focus of this research was on the advanced treatment train of the WTP (Figure 3.2). Commissioned in 2007, the advanced treatment train consists of OBM processes (ozonation, biologically active carbon filtration, ultrafiltration membranes) as well as chlorine disinfection. The OBM2 facility was added to WTP in July 2014 after this study.

Figure 3.1 The Lakeview WTP aerial photo. (Reprinted with permission from the Region of Peel, 2013)

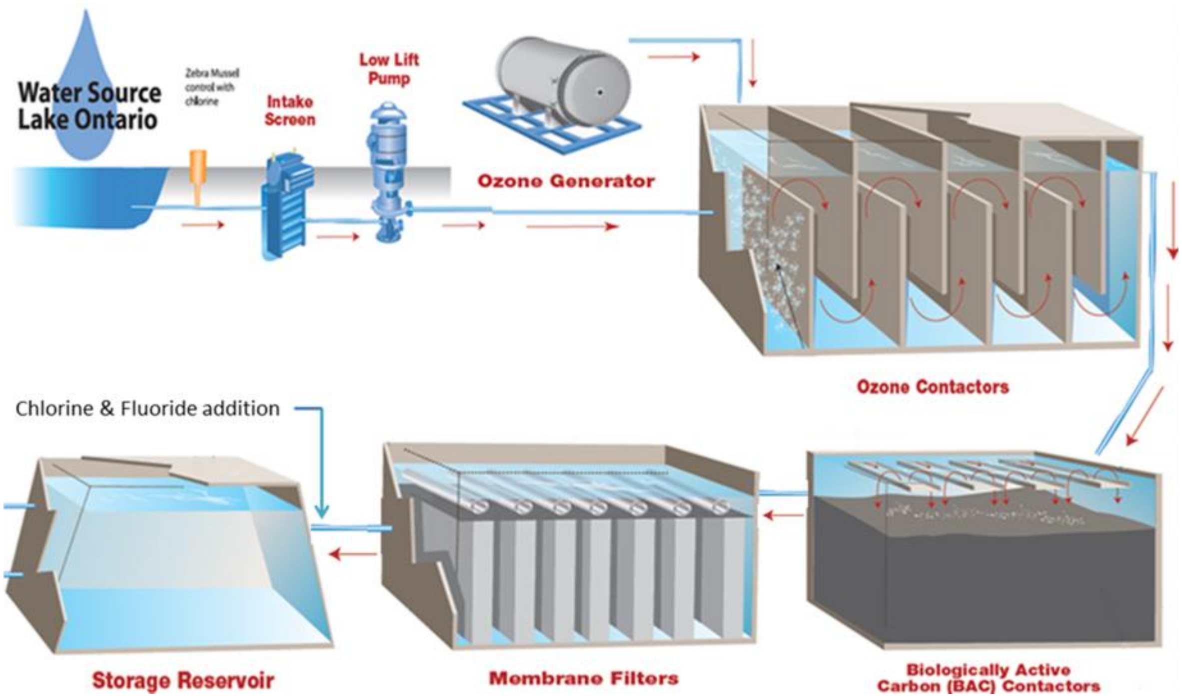

Figure 3.2 Schematic diagram of the advanced treatment train at the Lakeview WTP. (Reproduced with permission from the Region of Peel)

The Lakeview WTP draws water from Lake Ontario, which is of generally good quality with low total organic carbon (TOC) and dissolved organic carbon (DOC) concentration but occasional turbidity spikes and seasonal taste and odor episodes. Table 3.1 presents the raw water characteristics monitored biweekly from January to March 2013 and weekly from April 2013 to May 2014 (except for April 30, December 24 and 31 in 2013). Details can be found in Appendix D. The plant’s raw water was low in TOC and DOC concentration (~2.0 mg/L), alkalinity (<100 mg $CaCO_3$/L) and specific ultraviolet absorbance (SUVA) (~1 L/(mg C·m)) and relatively low in conductivity. In addition, the temperature of the raw water was quite low on average (7.9°C), which could have an impact on plant operation. Table 3.2 summarizes some important parameters on the Lakeview WTP finished water quality. Except for turbidity, which is monitored continuously, the other parameters are based on scheduled grab samples from January to December 2013. The WTP is able to effectively remove contaminants and produce water in compliance with Ontario's regulations.

Table 3.1 Lakeview WTP raw water characteristics

Parameters	Unit	Average	Minimum	Maximum
Temperature	°C	7.9	1.6	18.5
pH	-	7.61	7.08	7.99
Turbidity	NTU	0.47	0.24	1.64
Conductivity	µS/cm	318	255	403
Alkalinity	mg $CaCO_3$/L	89	75	102
TOC	mg C/L	2.06	1.61	2.69
DOC	mg C/L	2.01	1.59	2.60
SUVA	L/(mg C·m)	0.89	0.64	1.18

Table 3.2 Lakeview WTP finished water quality (Region of Peel 2013 Water Quality Report)

Parameters	Unit	Acceptable range/upper limit	Min-Max
Temperature	°C	15	1.8 – 22.2
pH	-	6.5 – 8.5	7.05 – 7.45
Turbidity	NTU	0.1	0.01 – 1.0*
Alkalinity	mg $CaCO_3$/L	30 - 500	86 - 220
Chloride	mg/L	250	10 - 30
Total dissolved solids	mg/L	500	160 - 229

*Turbidity results shown are from the advanced treatment train only, and turbidity is below 0.1 NTU 99% of the time.

3.2 Pre-chlorination

As raw water enters the Lakeview WTP intake, it is chlorinated for zebra mussel control. Water is then pumped into both treatment trains. Before the water enters the treatment facility, it passes through travelling screens to prevent items such as fish, shells and plants from entering. As soon as water reaches the advanced treatment train (OBM facility), sodium hydroxide and sodium bisulphite are added for pH control for de-chlorination, respectively.

3.3 Ozonation

The water then enters the ozone contactors. The ozonation process at the Lakeview WTP was designed for taste and odor control, but it also has the following advantages: disinfection, contaminant removal, increasing biodegradable organic matter (BOM) concentration by oxidizing organics, and UF membrane fouling control (Farr & Stampone, 2007). There are two ozone contactors which are designed to share the flow equally. Ozone is generated on-site from liquid oxygen with high voltage electricity and added to water through fine bubble diffusers located in the first two cells at the beginning of each ozone contactor. The ozone contactors are each designed for an effective contact time of approximately 18 minutes at a flow of 230 ML/d, which is theoretically sufficient to provide 1-log inactivation of *Cryptosporidium*. Ozone dosage ranged from 1.5 to 2.2 mg/L assuming a pH of 8 and average summer (15°C) and winter (8°C) conditions, respectively. Excess ozone accumulating in the head space of the contactor is removed and destroyed by thermal catalytic destructors before being exhausted to the atmosphere. Any residual ozone concentration remaining in the water is quenched with sodium bisulphite before it exits the ozone contactors. It is intended that the water leaving the ozone contactors be both ozone and chlorine free (Region of Peel, 2011).

3.4 Biologically Active Carbon Contactors

The water then passes into five biologically active carbon contactors (BACCs). The BACCs are rapid biofiltration units running in down mode (water passes vertically) and declining rate mode. Flow rate is dependent on the plant water demand. The BACCs are filled with granular activated carbon (GAC) with a bed depth of 2.5 m in each contactor. There is 2 m of water above the media during normal operation, and the volume of each contactor is 245.3 m^3. The specifications of the GAC used in the BACCs at the Lakeview WTP are presented in Table 3.3.

Table 3.3 Specifications of GAC used in the BACCs (Material Fact Sheet, Calgon Carbon)

GAC Parameters	Type	Effective Size	Uniformity Coefficient	Apparent Density
Value	Calgon FILTRASORB® 816	1.3 – 1.5 mm	1.4	0.55 g/cc

The BACCs are designed to operate in biological mode since the effluent of ozone contactors is chlorine and ozone free. The GAC is intended to provide habitat for microorganisms to attach and grow. The average viable biomass quantity as measured by adenosine triphosphate (ATP) throughout this study (January 2013 – May 2014) was 150 ng ATP/cm^3 dry GAC. Biological processes in the BACCs can remove excess BOM formed during ozonation thereby reducing microbial regrowth potential in distribution systems. In addition, the BACCs are able to remove turbidity and certain fractions of natural organic matter (NOM) to reduce both colloidal and organic fouling in downstream UF membranes. The average turbidity removal efficiency by the BACCs was 62%. Detailed data on NOM removal and fouling control by the BACCs is presented and discussed in subsequent chapters. The accumulation of solids and biological material will lead to headloss of the biofilters. Similarly to dual media filters, headloss development in the BACCs is controlled by periodic backwash. Each BACC is backwashed approximately every 120 operating hours. The backwash process involves a combination of water rinse and air scour.

3.5 Ultrafiltration

Effluent of the BACCs then passes through the ultrafiltration membranes (Figure 3.3). The UF membranes effectively remove suspended solids that were not removed by the upstream BACCs as well as pathogens. The UF membranes must have a log removal value (LRV) of ≥ 4.0 calculated from the membrane integrity test to be put into service and they are able to achieve an average 90% turbidity removal. The effluent of the advanced treatment train is consistently of high quality with an average turbidity of 0.01 NTU. The Lakeview WTP utilizes ZeeWeed® 1000 UF membrane modules, which consist of thousands of hollow fibres. The properties of the membrane module are shown in Table 3.4.

Figure 3.3 Membrane effluent treatment pipes at the Lakeview WTP

Table 3.4 ZeeWeed® 1000 Membrane Module Properties (ZeeWeed® 1000 Fact Sheet, GE Water & Process Technologies)

Material	Nominal Pore Size	Nominal Surface Area	Surface Properties	Fibre Diameter	Flow Path
PVDF	0.02 µm	46.5/55.7* m^2	Non-ionic & hydrophilic	OD: 0.95 mm ID: 0.47 mm	Outside-in

*Modules installed in 2007 have nominal surface area of 46.5 m^2 (500 ft^2); modules installed in 2009 have nominal surface area of 55.7 m^2 (550 ft^2).

The membrane modules are placed horizontally and assembled into cassettes, which are submerged in 12 membrane tanks (trains). Once water is directed into these membrane tanks, the negative pressure on membranes created by permeate pumps pulls water from outside the membranes vertically through the fibre walls. The UF membrane system is operated as a simple semi-batch process where filtration and backwash alternate in sequence. During filtration, a constant level in the membrane tank is maintained by replacing permeate with feed water. Flux of the membranes is dependent on plant water demand, and therefore it may vary considerably (±10%). Flux through each membrane train is adjusted to minimize overall feed flow fluctuations. At the end of each filtration cycle which typically lasts for 42 minutes, a backwash is performed typically for 30 seconds. During backwash, the membranes are simultaneously aerated and backpulsed to dislodge solids from the membrane surfaces and pores. Once the backwash process is complete, the tank is completely drained with aeration, which rids the tank of any accumulated solids. The tank is then refilled with feed water and production resumes (Region of Peel, 2011).

As hydraulically irreversible fouling develops, backwashing ultimately cannot clean the membranes sufficiently. In order to maintain the flux required for production under constant pressure, chemical cleaning is used to remove resistant deposition on membrane surfaces and within pores. At the Lakeview WTP, a maintenance clean is performed on each membrane train following each 33 ML volume of water produced. It involves recirculating permeate with 100 mg/L sodium hypochlorite in the membrane tank for 30 minutes. The membrane tank is then drained with aeration. Recovery cleaning involves higher concentrations of cleaning agent and longer contact time. A recovery clean using 250 mg/L sodium hypochlorite is performed every 42 days, and a citric acid recovery clean with 1 g/L is performed twice a year, both with a 6 hour recirculation time. Heated water is used when the water temperature is below 10°C.

3.6 Chlorination

Before effluent of the ozonation-biofiltration-membrane (OBM) process and treated water from the conventional treatment train are directed to a storage reservoir, the water is temporally stored in a weir box. This is also where chlorine is added for secondary disinfection. A 0.5 log inactivation of *Giardia* is targeted through chlorination. Fluoride addition occurs in the weir box as well. Treated water from the conventional treatment train and advanced treatment trains is mixed in the storage reservoir, and the final finished water is ultimately pumped from the storage reservoir into distribution systems.

Chapter 4 Biofiltration with and without Ozone as an Ultrafiltration Membrane Pre-treatment

Summary

Fouling remains one of the major constraints on the application of low pressure membranes (LPMs) in drinking water treatment. Recent studies have shown that biofiltration is able to reduce fouling in LPMs. The purpose of this study was to investigate the effect of ozonation prior to biofiltration as an integrated LPM pre-treatment. The investigation was carried out at a full-scale water treatment plant which employs ozonation, biofiltration and ultrafiltration (UF). Additionally, a UF pilot plant was operated with effluent from a full-scale biofilter. The operation of ozone was observed to have a significant impact on biomass activity within the biofilters. Ozone was able to reduce specific UV absorbance (SUVA) and decrease the retention of biopolymers by both full-scale and pilot-scale UF membranes. Biopolymers were implicated as key organic foulants. With ozone online, hydraulically reversible and irreversible fouling rates were 50% lower than with ozone out of service.

4.1 Introduction

Fouling remains a major challenge for the further adoption of membrane processes in drinking water treatment. Membrane fouling is caused by the accumulation of material on membrane surfaces and within membrane pores; it leads to increases in operational complexity and maintenance costs, deterioration in productivity and shorter membrane service life (Crittenden *et al*., 2012). Membrane fouling can be controlled by backwashing, chemical cleaning and pre-treating the feed water. Extensive studies have focused on the selection, mechanisms, and optimization of appropriate pre-treatment processes to control membrane fouling.

Natural organic matter (NOM) can be an important type of foulant, especially for low pressure membranes (LPMs) (Howe & Clark, 2002; Amy, 2008; Cai & Benjamin, 2011). Using microfiltration (MF) and ultrafiltration (UF) systems to treat different natural waters, Howe & Clark (2002) found that fouling in low pressure membrane systems was predominantly caused by organic colloids (hydraulically reversible or irreversible fouling not specified). Lee et al (2004) confirmed this finding and further concluded that polysaccharides and proteins are significant organic foulants among different NOM components. More recently with the development of high performance size exclusion chromatography (HP-SEC), the importance of NOM fractions and molecular weight distribution on LPM fouling have been recognized. For example, with the assistance of liquid chromatography-organic carbon detection (LC-OCD), biopolymers, which consist of polysaccharides and protein-like material, have been identified as the critical fraction of NOM responsible for organic fouling (Hallé *et al.*, 2009; Peldszus *et al.*, 2011; Peldszus *et al.*, 2012).

Ozonation is reasonably commonly used in water treatment, and its application in membrane systems is promising because ozonation pre-treatment can reduce downstream membrane fouling by altering the composition and size distribution of NOM (Van Geluwe *et al.*, 2011). Hashino et al (2000 & 2001) used ozone-resistant MF membranes following ozonation for drinking water production, and found that the permeate flux was three to four times higher with pre-ozonation than without it. The authors also concluded that the reaction of ozone with DOM has to occur on the surface of the membrane to have such effect. In an investigation on an ozone-UF hybrid system, the researchers attributed flux enhancement to change in particle size distribution and membrane zeta potential (Hyung *et al.*, 2000). To investigate NOM fouling in a hollow fibre MF system, Song et al (2010) used apparent molecular weight distribution (AMWD) to characterize NOM. The authors concluded that large molecular weight organic matter is responsible for most of the fouling, and ozonation can change the AMWD and reduce fouling. Also consistent with previous studies is that ozone oxidizes the more hydrophobic fractions more than hydrophilic ones, and membrane performance improves with increasing ozone concentration until a certain dosage. In the Song et al (2010) study, the optimal ozone dose was 1.5 mg/L in the 0.5 - 3.0 mgO_3/L range. Similar findings were made in water reuse applications (Wang *et al*., 2007; You *et al*., 2007) as well as in ceramic membranes, which are less vulnerable to ozone damage (Karnik *et al*., 2005a; Kim *et al*., 2008).

In drinking water treatment, it is common practice to follow ozonation with biological filtration to reduce the excess biodegradable organic matter (BOM) induced by ozonation in order to control microbial regrowth in distribution systems. However, very few studies have investigated the possibility of using ozonation combined with biofiltration as a membrane pre-treatment (i.e., OBM process), not to mention its fouling control effectiveness and mechanisms. Nishijima & Okada (1998) first studied the particle separation capacity of ozonation combined with biological activated carbon (BAC) filtration and explored its opportunities as a pre-treatment for advanced water treatment. The authors concluded that significant levels of both particulate and dissolved organic matter were removed by the ozonation-biofiltration process, which could contribute to membrane fouling reduction. However, the experiment did not include a membrane system following the ozonation-biofiltration process to prove it. Osterhus et al (2007) investigated the particle removal capacity of the OBM process using NOM containing surface water and hollow fibre UF membranes. Though the focus of that study was not on fouling, the authors did observe differences in fouling rate at different pH values, which was attributed to different ozonation reaction pathways and by-products. However, the study was not able to characterize the change in NOM make-up during the process. Geismar et al (2012) thoroughly examined the fouling control capacity of ozonation and BAC filtration as LPM pre-treatments. The impacts of ozone dosage and membrane type were also investigated. The findings were consistent with studies with ozonation alone as a LPM pre-treatment in terms of fouling reduction, optimal ozone dosage and membrane type. However, the experiments were conducted at bench-scale only for days. As such, hydraulically irreversible fouling was too low to be detected. In addition, the authors treated ozonation and BAC filtration separately and were not able to investigate the interaction between the two processes.

In the present study, a long-term detailed investigation into ozonation-biofiltration as an integrated LPM pre-treatment was undertaken on a relatively large scale. The research focused on the role of ozonation in the integrated pre-treatment process, its fouling control mechanisms, and the interaction between ozonation and biological filtration. This study specifically assessed the fouling reduction capacity of ozonation-biofiltration and biofiltration alone as UF membrane pre-treatment options, and investigates the impact of ozone on biological activity within the biofilters. New insights into NOM transformation through these processes are also provided.

4.2 Materials and Methods

4.2.1 Lakeview Water Treatment Plant

This study was conducted from January 2013 to May 2014 at the Lakeview Water Treatment Plant (WTP) owned by the Region of Peel in southern Ontario. The plant draws water from Lake Ontario. The raw water characteristics for the study period were previously discussed in Chapter 3 and are presented again in Table 4.1. At the time of this research, the WTP had a capacity of 800 ML/d and consisted of a conventional treatment train and an advanced treatment train. The focus of this investigation was on the advanced treatment train of the plant. The advanced treatment train proessed half of the plant capacity (i.e., 400 ML/d) and employed OBM processes (i.e., ozonation, biologically active carbon filtration, ultrafiltration membranes) as well as chlorine disinfection. An OBM2 facility was added to the Lakeview WTP in July 2014 following the completion of this study. In the OBM1 facility, where this study was conducted, there are 5 parallel biologically active carbon contactors (BACCs) and 12 membrane trains. During this study, ozone was out of service from January to March 2013, May to June 2013, and January to March 2014, which allowed for direct comparison on biological activity within the BACCs, UF membrane fouling rate and NOM composition with and without the effect of ozonation. When ozone was online, dosages ranged from 1.5 to 2.2 mg/L determined at a pH of 8 and average summer (15°C) and winter (8°C) water conditions, respectively (Region of Peel, 2011).

Table 4.1 Lakeview WTP raw water characteristics from January 2013 to May 2014 (n=58)

Parameters	Unit	Average	Minimum	Maximum
Temperature	°C	7.9	1.6	18.5
pH	-	7.61	7.08	7.99
Turbidity	NTU	0.47	0.24	1.64
Conductivity	µS/cm	318	255	403
Alkalinity	mg $CaCO_3$/L	89	75	102
TOC	mg C/L	2.06	1.61	2.69
DOC	mg C/L	2.01	1.59	2.60
SUVA	L/(mg C·m)	0.89	0.64	1.18

4.2.2 UF Pilot Plant

A UF membrane pilot plant (ZeeWeed®10, GE Water and Process Technologies, Oakville, ON, Canada) was fed with the effluent from BACC#3. The pilot plant contained a polyvinylidene fluoride (PVDF) hollow fibre ZeeWeed®500 membrane module with a nominal surface area of 1.0 m^2 immersed in a 15 L membrane tank. A ZeeWeed®500 membrane module is similar to the ZeeWeed®1000 module employed at the Lakeview WTP in many aspects including material (PVDF), flow path (outside-in), and surface properties (non-ionic & hydrophilic). However, the ZeeWeed®1000 module has thinner fibres and smaller nominal pore size than ZeeWeed®500, which has a nominal pore size of 0.04 µm (data provided by the manufacturer). The pilot unit was programmed to continuously monitor and record data on temperature, flow, permeability, and trans-membrane pressure (TMP) with its built-in temperature sensor, flow meter and pressure transducer.

The pilot unit was operated in dead-end mode at constant permeate flux with periodic backflushing. Water in the membrane tank was drawn by a vacuum pump through fibre walls (outside-in) and ultimately into a permeate tank. A typical operation sequence included (1) permeation for 30 min, (2) back pulse with aeration for 30 s, (3) aeration for 15 s (4) draining of the tank with aeration, and (5) filling of tank for 2 min. The unit was operated from May 2013 to May 2014 with a permeate flux of 52 $L/m^2/h$ (LMH) except from December 17 2013 to January 6 2014 when it was operated at a flux of 26 LMH without human supervision and maintenance cleaning. The membrane module was new at the start of operation. Except for the period mentioned, maintenance cleaning was performed manually twice a week (typically on Tuesdays and Fridays) by soaking the membrane in 100 mg/L sodium hypochlorite solution for 20 min. Membrane integrity tests and clean water permeability tests were conducted regularly to ensure sound membrane condition and effectiveness of cleaning. Fibre repairs were performed on a few occasions as required.

4.2.3 Sample Collection and Analyses

Water sampling was conducted at the Lakeview WTP biweekly from January to March 2013 and weekly from April 2013 to May 2014 (except for April 30, December 24 and 31 in 2013). As shown in Figure 4.1, water samples were taken essentially after each treatment process including raw water after intake screen, ozone effluent, effluent of BACC#3 and BACC#5, permeate of UF pilot plant, and permeate from one of the full-scale membrane trains (UF#42).

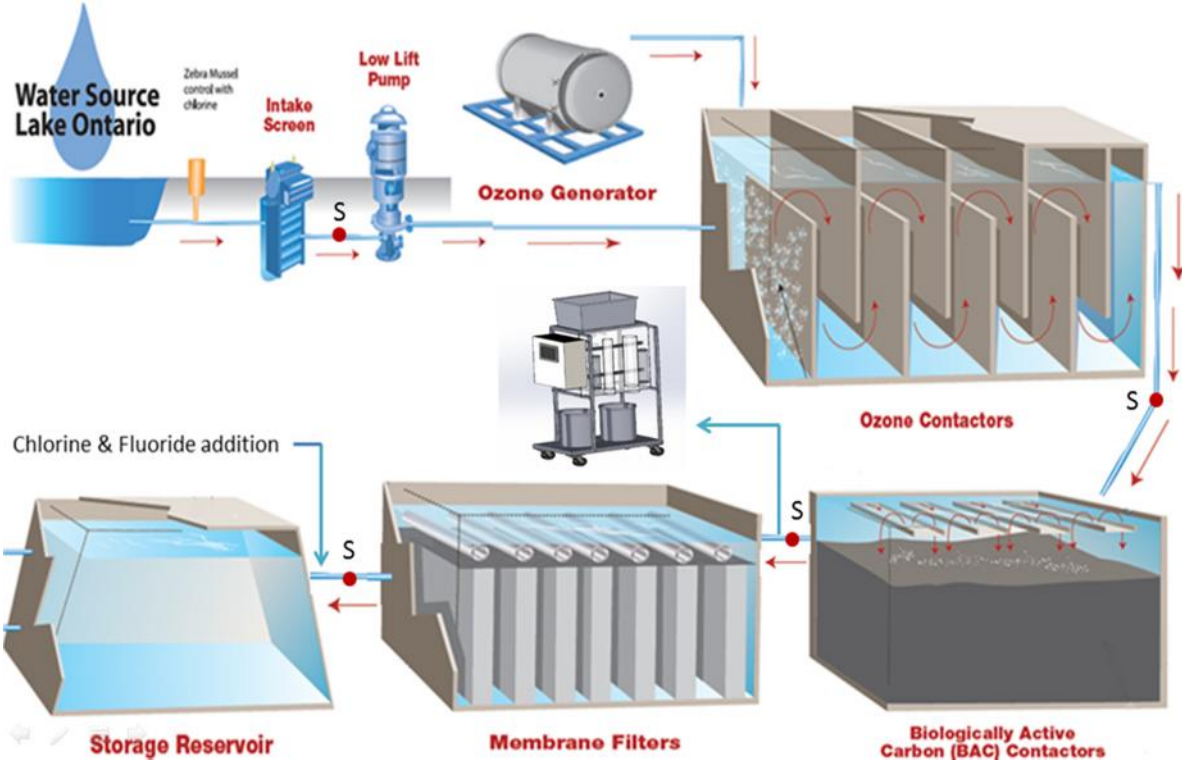

Figure 4.1 Lakeview WTP advanced treatment train process schematic with UF pilot plant and sampling locations (S represents sampling locations). (Reproduced with permission from the Region of Peel)

The following water quality parameters were measured in some or all of the samples taken: pH, temperature, alkalinity, conductivity, turbidity, chlorine residual, UV_{254}, and specific UV absorbance (SUVA) using standard methods. Total and dissolved organic carbon (TOC/DOC) were measured using an OI-Analytical TOC analyzer model 1010 (College Station, TX, USA) from January to August 2013 and model 1030 from August 2013 to May 2014 by wet-oxidation method (Standard Methods 5310D). TOC/DOC samples were filtered through 0.45μm polyethersulfone filters and preserved with phosphoric acid to pH 2 before measurements. UV_{254} was measured with UV-Vis spectrometer (Cary 100, Agilent Technologies, Mississauga, ON), and SUVA was calculated as follows: SUVA = UV_{254}/DOC. Liquid chromatography with organic carbon detection (LC-OCD) (DOC-Labor Dr. Huber, Karlsruhe, Germany) analysis was employed to measure the concentrations of the various NOM fractions including biopolymers, humic substances, building blocks, low molecular weight (LMW) acids and humics, and LMW neutrals in water as described in Huber et al (2011). Total chlorine residual was measured onsite using a pocket colorimeter^{TM} II (Hach, Loveland, CO). Turbidity was monitored using online turbidimeters (Hach 1720E and FT660sc). Other parameters were analyzed at the University of Waterloo laboratories, and if not measured on-line, samples were stored at 4°C until measurements.

4.2.4 Biomass Analyses

Granular activated carbon (GAC) samples were collected from the top of BACC#3 biweekly to analyze biomass activity within the BACCs. Adenosine triphosphate (ATP), an indicator of viable biomass, was measured from February 2013 to May 2014 using a LuminUltra™ DSA ATP test kit (LuminUltra, Fredericton, NB, Canada). Total ATP was calculated by measuring the relative light unit (RLU) of extracted media with a luminometer (Modulus Luminometer, model 9200-102, Turner BioSystems, Sunnyvale, CA). In addition, fluorescein diacetate (FDA) measurements were performed from May 2013 to May 2014 to quantify overall microbial activity using a method adapted from Green et al (2006). FDA can be hydrolyzed by important enzymes (proteases, lipases and esterase) in microbial metabolism and fluorescein is a product of FDA hydrolysis. The amount of fluorescein produced is proportional to the amount of active enzymes within the biomass (Adam & Duncan, 2001; Green *et al*., 2006). Briefly, the process involved a 3 h incubation of 1g wet GAC sample in 50 mL sodium phosphate buffer with 0.5 mL of 4.9 mM FDA substrate at 37°C. During incubation, FDA was hydrolyzed, and fluorescein was released. After incubation, samples were centrifuged for two minutes. The fluorescein was then measured with a spectrophotometer at 490 nm and compared with a fluorescein standard curve with the following concentrations: 0, 30, 100, 300 and 500 μg fluorescein per 50 mL. A blank made of 50 mL sodium phosphate buffer and 0.5 mL acetone without GAC sample was also used.

4.3 Results and Discussion

4.3.1 Particle Removal Efficiencies by Different Treatment Processes

The Lakeview WTP's raw water is generally cool to cold and relatively low in TOC and DOC (~2.0 mg/L), alkalinity (<100 mg $CaCO_3$/L), specific ultraviolet absorbance (SUVA) (~1 L/(mg C·m)), and conductivity (Table 4.1). There were occasional turbidity peaks during this study; even so, the maximum measured on our sampling days was only 1.6 NTU. However, the organic carbon concentration varied by only 14%. The TOC, DOC and turbidity removal efficiencies through the plant's OBM processes are presented in Table 4.2. The removal efficiencies presented are net removals with respect to the previous process, which take into account both the value removed and/or generated by a process (i.e. not cumulative with the exception of the O_3 + BACC column). Ozonation data and ozonation-biofiltration combined data presented in this table are from samples collected only when ozone was online, whereas BACC and UF data include all samples. Student t-tests ($\alpha = 0.05$) confirmed that there was no significant difference in BACC and UF removal efficiencies when ozone was online and out of service with respect to TOC/DOC and turbidity.

Table 4.2 Average percentage removal and standard deviation by treatment processes

Process	O_3	BACC	O_3 + BACC	UF
TOC	4 (±3)	3 (±3)	7 (±4)	12 (±5)
DOC	3 (±4)	3 (±3)	6 (±4)	12 (±6)
Turbidity	56 (±17)	59 (±15)	73 (±15)	90 (±6)

The ozonation and BACC processes individually and in combination achieved minimal organic carbon removal. However, ozonation at the Lakeview WTP was not designed to remove TOC/DOC. Its main function is to control taste and odor. Although it would be expected that ozone can provide some disinfection, this is not taken into consideration for regulatory purpose. As is well known, ozone can react with organic carbon and change the characteristics of natural organic matter (NOM), but complete mineralization is rarely achieved under typical water treatment ozone dosages. Therefore, the net removal of TOC/DOC by ozonation was minimal. One important reason for the low organics removal by the BACCs at the Lakeview WTP was considered to be the low water temperature. The annual average was around 8°C, and the temperature never exceeded 20°C during the study. Temperature is of course an influential factor in biological processes with low temperatures reducing microbial activity. For example, Emelko et al (2006) observed significant difference in organics removal by biofilters between warm (21-24°C) and cold (1-3°C) conditions. Secondly, although biofilters can remove certain NOM, some organic by-products can also be produced by biofilters, such as soluble microbial products (SMPs), which could contribute to the low net organics removal by the BACCs. About 12% of the organic carbon was retained by the UF membranes. Since the UF membranes are designed to remove particles and pathogens not organics, this means that 12% of the TOC/DOC constituents are potential membrane foulants. The identification of these foulants is discussed later in this chapter.

The turbidity of water at different stages in the OBM treatment train is presented in Figure 4.2. Turbidity data for ozone contactor effluent were only available from August 2013 on, and includes measurements from both when ozone was online and out of service. Both the ozonation and biofiltration processes were generally able to remove over 50% of the influent turbidity, individually. In combination, the ozonation-biofiltration pre-treatment process removed on average 73% of the turbidity in raw water before it reached the downstream UF membranes. Sedimentation which occurred within the ozone contactors (dissipation tanks) contributed to some removal of turbidity. The average turbidity removal occurring in the ozone contactors was 56% with ozone online and 41% when ozonation was out of service as water continued to pass through the ozone contactors. The BACCs achieved a turbidity removal rate of 59% based on ozone contactor effluent. The UF membranes achieved high and consistent turbidity removal, with the average membrane permeate turbidity being 0.01 NTU.

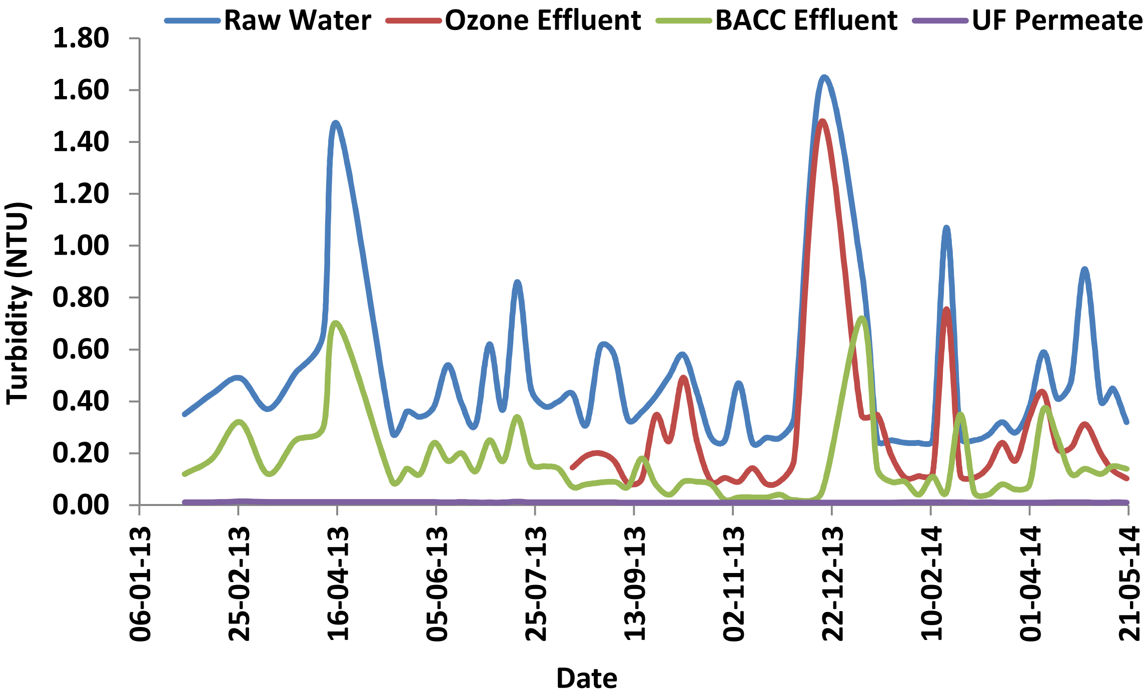

Figure 4.2 Turbidity following different treatment stages

4.3.2 The Effect of Ozone on Biomass in the BACCs

Adenosine triphosphate (ATP) method is commonly used to evaluate the viable biomass within drinking water biofilters as ATP is the primary energy carrier in all types of living cells and is used for cell synthesis and maintenance (Rittmann & McCarty, 2001). The overall average viable biomass at the Lakeview WTP as measured by ATP was 150 ng ATP/cm^3 dry GAC, which is on the lower end of the 100-1,000 ng ATP/cm^3 media range for typical acclimated biofilters (Pharand *et al.*, 2014). One possible explanation for the low viable biomass in the BACCs at the Lakeview WTP is the persistently low water temperature. The annual average of 8°C is not ideal for rapid and sustained development of biomass. The average temperature was 10°C with ozone online and 5°C with ozone out of service. A second possible reason may be the low organic carbon concentrations in the raw water. Organic carbon is the primary energy source for microorganisms; insufficient organic carbon could limit microbial growth. In addition, frequent changes in ozone application and operation were likely detrimental to continuous development and maintenance of viable biomass.

Fluorescein diacetate (FDA) method was used to evaluate overall biomass activity. FDA results are in proportion to important enzymes involved with microbial metabolism. The average biomass activity at the surface of the BACCs at the Lakeview WTP as measured by FDA was 99 µg fluorescein/cm^3 dry GAC. Results of biomass analyses from the GAC samples collected from BACC#3 are presented in Figure 4.3 using the surrogates ATP and FDA to quantify viable biomass and overall microbial activity, respectively. ATP and FDA results followed the same trends, which indicated that most of the viable biomass within the BACCs at the Lakeview WTP was active. In addition, both methods were sensitive and responded quickly to changes. ATP and FDA data validated trends observed for both.

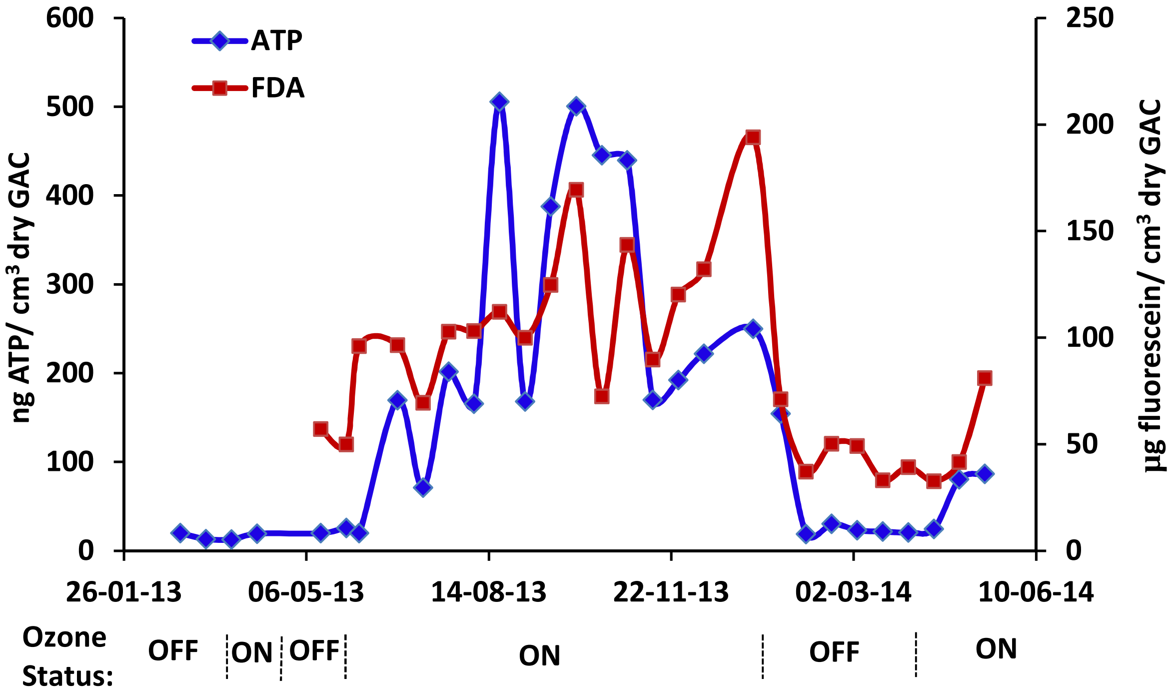

Figure 4.3 Biomass analyses from BACC#3 using the surrogates ATP and FDA

From Figure 4.3, it is evident that the operation of ozone had a substantial impact on both biomass quantity and activity. Student t-tests (α = 0.05) showed that both viable biomass and overall biomass activity were significantly higher with ozone online than out of service. The average ATP and FDA values when ozone was online were 205 ng ATP/cm^3 dry GAC and 114 µg fluorescein/cm^3 dry GAC, respectively; whereas with ozone out of service, the values decreased to 53 ng ATP/cm^3 dry GAC and 71 µg fluorescein/cm^3 dry GAC. This finding is consistent with Magic-Knezev and van der Kooij (2004), where the authors found that the ATP concentration in full-scale GAC biofilters with pre-ozonation was 2 or 3 times higher than biofilters under similar operating conditions fed with non-ozonated water. It is well-known that ozone can react with NOM. Specifically, ozone attacks double bonds and aromatic structures and breaks down large organic molecules present in water resulting in an increase in the concentration of biodegradable organic matter (BOM) (van der Helm *et al*., 2009). This will prompt biodegradation as well as microbial growth in biofilters.

4.3.3 The Effect of Ozone on NOM Composition

Specific UV absorbance (SUVA) is a water quality test parameter commonly used to indicate the hydrophobicity of dissolved organic matter. During this investigation, the raw water of the Lakeview WTP had an average SUVA value of 0.89 L/(mg C·m) ranging from 0.64 to 1.18 L/(mg C·m) (Figure 4.4). This low SUVA value was indicative of low hydrophobicity and low molecular weight content (less humic content) (Edzwald & Tobiason, 1999). When ozone was online, it was able to reduce SUVA by 42.8% on average. No other process was demonstrated to significantly reduce SUVA as per student t-test (α = 0.05). In fact, when ozone was out of service, SUVA values were consistent with raw water SUVA value at all treatment stages. This is expected because neither biofiltration nor UF membrane is able to substantially remove hydrophobic organic matter. Double bonds and aromatic structures are very difficult for microorganisms in biofilters to biodegrade, and UF membranes can only remove macromolecules, such as proteins and polymers. SUVA can also be indicative of biodegrability (Juhna & Melin, 2006), who reported that low SUVA values indicate high biodegrability. In the present study, ozonation substantially reduced SUVA, which would be another indication that ozonated water was more biodegradable. This is in alignment with the biomass quantity and activity response to ozone operation discussed earlier.

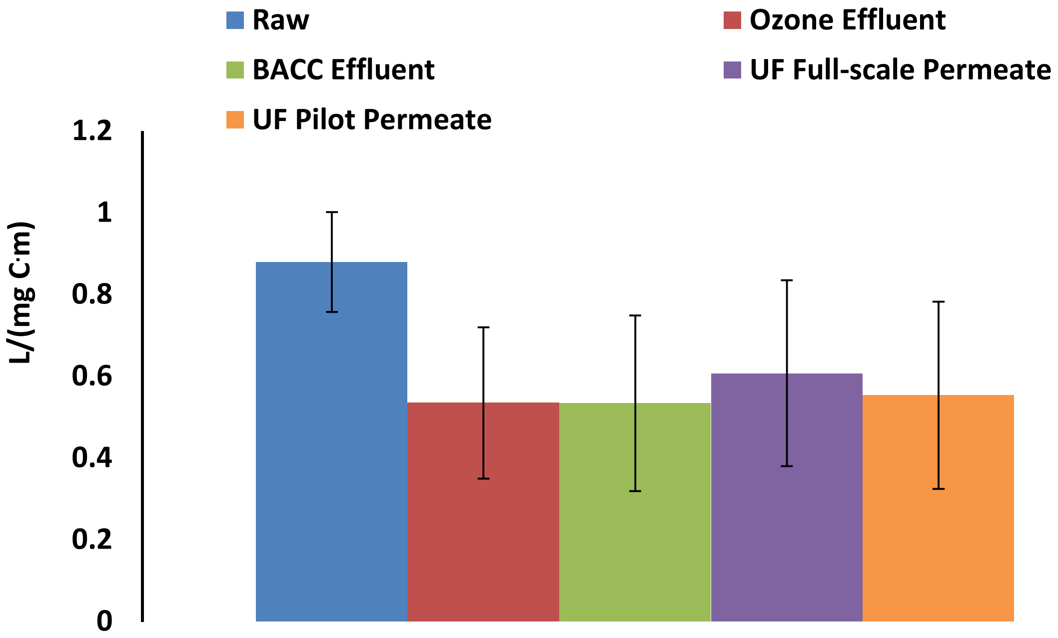

Figure 4.4 Average SUVA values at following different treatment stages during ozone online (n=33). Error bars show standard deviation.

DOC and SUVA can provide information on the characteristics of NOM, whereas high performance size exclusion chromatography (HP-SEC), such as liquid chromatography-organic carbon detection (LC-OCD) is able to provide insights on the composition of NOM. During this investigation, the DOC in incoming raw water at the Lakeview WTP contained 13.3% biopolymers, 49.6% humic substances, 18.8% building blocks, and 10.4% low molecular weight (LMW) neutrals based on 50 LC-OCD measurements. The LMW acids and LMW humics concentration constituted less than 1% of DOC on most occasions, and the remaining 7.9% did not fall into these fractions. The concentrations of different NOM fractions at different stages of the OBM treatment train at the Lakeview WTP during ozone online are presented in Figure 4.5.

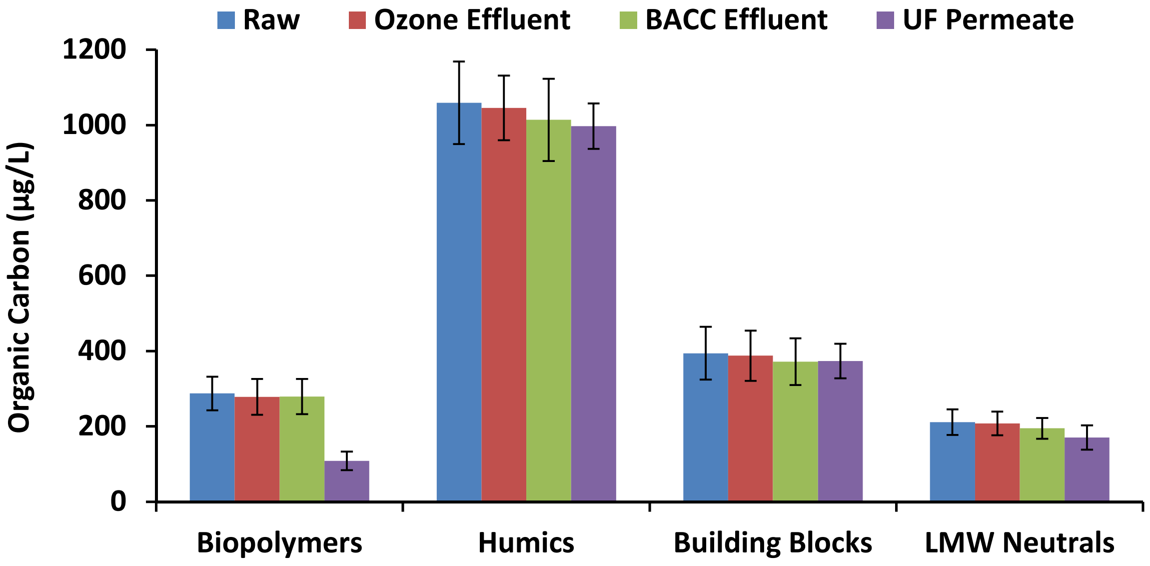

Figure 4.5 Concentrations of different NOM fractions at following different treatment stages during ozone online (n=38). Error bars show standard deviation.

LMW acids and LMW humics concentrations were so low that they can essentially be neglected. Humic substances made up the largest fraction of the NOM, and this is consistent with previous studies which reported that humic substances typically represent 40 to 60% of the total DOC in natural waters (Thurman, 1985). There was minimal removal of humic substances by the OBM process, which is consistent with the very low DOC removal rates by ozonation and biofiltration. This also indicated that the humic substances present in raw water of the Lakeview WTP were not responsible for significant UF membrane fouling. This suggests biopolymers, which consist of polysaccharides and proteins, account for 13% of DOC in raw water. However, 61% of the biopolymer fraction was retained by the UF membranes, strongly suggests that biopolymers are the fraction of NOM responsible for membrane fouling. Retention is defined here as the net removal of a process. This finding is in line with previous membrane studies in which LC-OCD analysis was utilized for both drinking water (Her *et al*., 2007; Hallé *et al*., 2009; Peldszus *et al*., 2012) and membrane bioreactor applications (Jiang *et al*., 2010). However, in Hallé et al (2009) and Peldszus et al (2012) the biofiltration pre-treatment was able to effectively remove biopolymers, whereas the BACCs at the Lakeview WTP achieved minimal biopolymer removal. Contributing factors are the differences in the two types of waters investigated and experimental conditions. In Hallé et al (2009) and Peldszus et al (2012), the water investigated was a river water with a DOC concentration of 5-7 mg/L, which is two to three times higher than the lake water used in this investigation. In addition, the raw water temperature in those two studies was higher with a broader range (0-25°C). Those authors observed significant DOC and biopolymer removal by the biofilters under warm conditions (10-25°C), but no removal when the water temperature was below 2°C. These observations help to explain the minimal DOC and biopolymer removal by the BACCs in this study due to low temperature. Potential differences in the characteristics of the biopolymers may also play a role.

Though ozone substantially reduced SUVA, it did not significantly remove any of the NOM fractions. The NOM fractions measured by LC-OCD are classified based on apparent molecular weight (AMW), shape, and chemical interaction. There is no direct correlation between SUVA value and concentrations of the different NOM fractions. At typical drinking water applied doses, ozone only changes characteristics of NOM but does not significantly reduce its total concentration as mineralization rarely occurs (van der Helm *et al*., 2009;

Treguer *et al*., 2010). This was also the case for present study. Ozone has been found to be able to change the apparent molecular weight distribution (AMWD) of NOM (Song *et al*., 2010; Geismar *et al*., 2012). However, the results produced by LC-OCD are not transferrable to AMWD. Pharand (2014) also performed LC-OCD analyses for samples from a full-scale WTP that employs ozonation. Our finding was consistent with Pharand (2014) in that ozone did not significantly remove any of the NOM fractions.

What is more important is the impact of ozone on membrane fouling and biopolymers, which has been shown to be the only fraction of NOM responsible for fouling for the water investigated. Biopolymer concentrations at different stages of the OBM treatment train at the Lakeview WTP are presented in Figure 4.6, and comparison between ozone online and out of service is made. Interesting though, ozone did not remove any of the NOM fractions, it did have an impact on biopolymer retention by the UF membranes. With ozone online, fewer biopolymers were retained by the UF membranes, which likely leads to lower fouling rates.

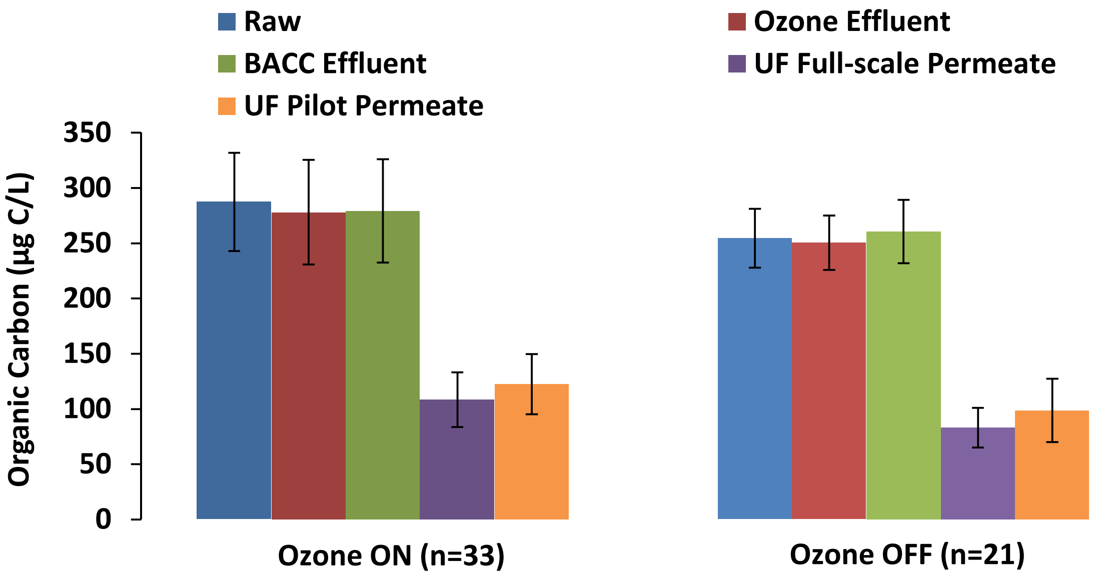

Figure 4.6 Comparison of biopolymer concentrations at different treatment stages between ozone online and out of service. Error bars show standard deviation.

During the 16 month investigation, ozone was out of service from January to March 2013, May to June 2013, and January to March 2014. Over 30 LC-OCD measurements were performed including both permeate from full-scale UF membranes and membrane pilot plant with ozone online and 21 measurements with ozone out of service. Average biopolymer concentrations in raw water, membrane permeate, and biopolymer retention rates by both full-scale and pilot-scale UF membranes with ozone online and offline are shown in Table 4.3. The average biopolymer retention by full-scale and pilot-scale membranes was 7% and 9% lower with ozone online than ozone offline, respectively, the difference being statistically significant as per student t-tests ($\alpha = 0.05$). Therefore, it can be concluded that ozone was able to decrease biopolymer retention by UF membranes.

Table 4.3 Average biopolymer concentrations and retention (with standard deviation) by full-scale and pilot-scale membranes during period of ozone online and out of service

		UF Full-scale		UF Pilot-scale	
	Raw water concentration (µg/L)	Permeate concentration (µg/L)	Retention (%)	Permeate concentration (µg/L)	Retention (%)
O_3 on	288 (±44)	108 (±24)	61 (±5)	122 (±27)	56 (±8)
n	38	38		33	
O_3 off	255 (±27)	83 (±18)	68 (±6)	99 (±29)	65 (±6)
n	21	21		17	

n = number of samples

It can be further concluded that ozone can alter the interactions between foulants and membrane surfaces. The most important interaction between foulants and membrane surface is size exclusion, which refers to the rejection of particles by membranes based on size (AWWA, 2005). Based on the results from this investigation as well as findings from previous studies on the impact of ozone on AMWD, it can be inferred that ozone is able to reduce the average size of biopolymers. As a result, more biopolymers (foulants) can pass through UF membranes. This is very likely because ozone is known to be able to break down large organic molecules, and biopolymers are the fraction of NOM with the highest molecular weight (von Gunten 2003; Huber *et al*., 2011).

4.3.4 The Impact of Ozone on UF Membrane Fouling

The UF pilot unit was operated from May 2013 to May 2014 and fed with effluent from full-scale BACC#3. During this period, ozone was out of service from May to June 2013 for 4 weeks and from January to April 2014 for 12 weeks. The pilot plant was operated at 52 $L/m^2/h$ (LMH) except for three weeks from December 2013 to January 2014 when it was run at 26 LMH. Maintenance cleaning was performed twice per week. Fibre repair was performed once as the membrane module did not pass the integrity test. Since the membrane was operated at constant flux, the build-up of trans-membrane pressure (TMP) should approximate fouling rate. TMP data after each membrane backpulse, captured by the pilot unit for the complete period of operation, is shown in Figure 4.7, and temperature corrected TMP (at 20°C) before and after membrane backpulse is shown in Figure 4.8. Temperature correction was calculated based on the following formula:

$J (T) = J (20) * 1.025^{(T-20)}$

Where J (T) is the temperature corrected flux at the anticipated temperature T (°C), and J (20) is the flux at 20°C. This is an empirical equation derived from correlation between temperature and viscosity.

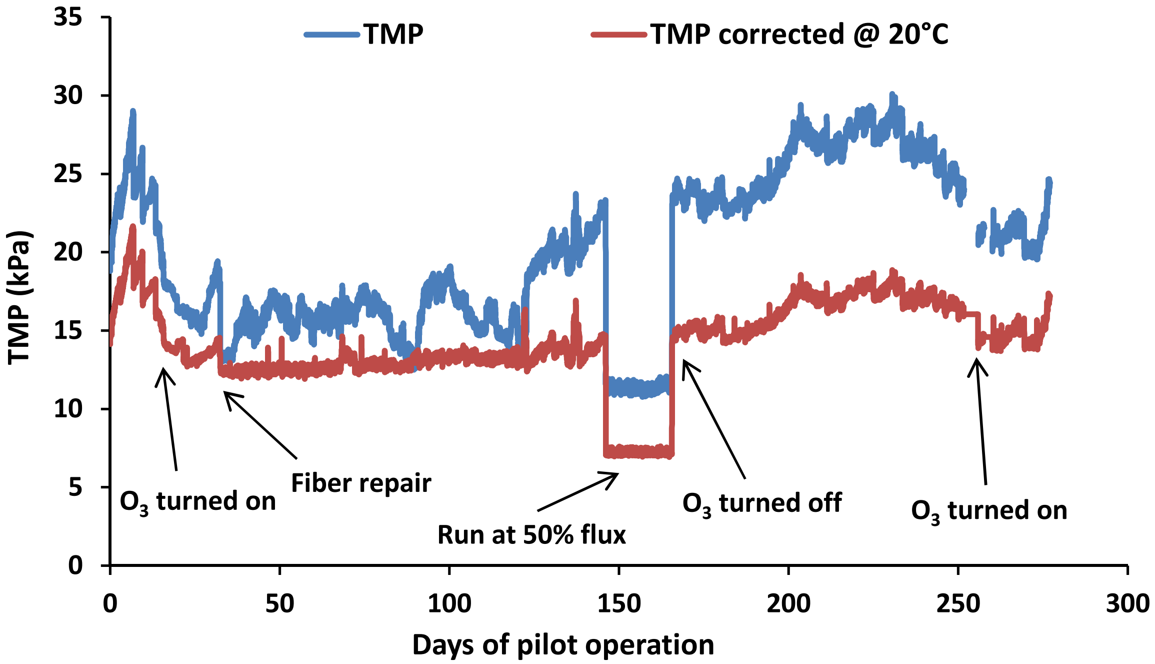

Figure 4.7 Pilot plant complete TMP data set after backpulse

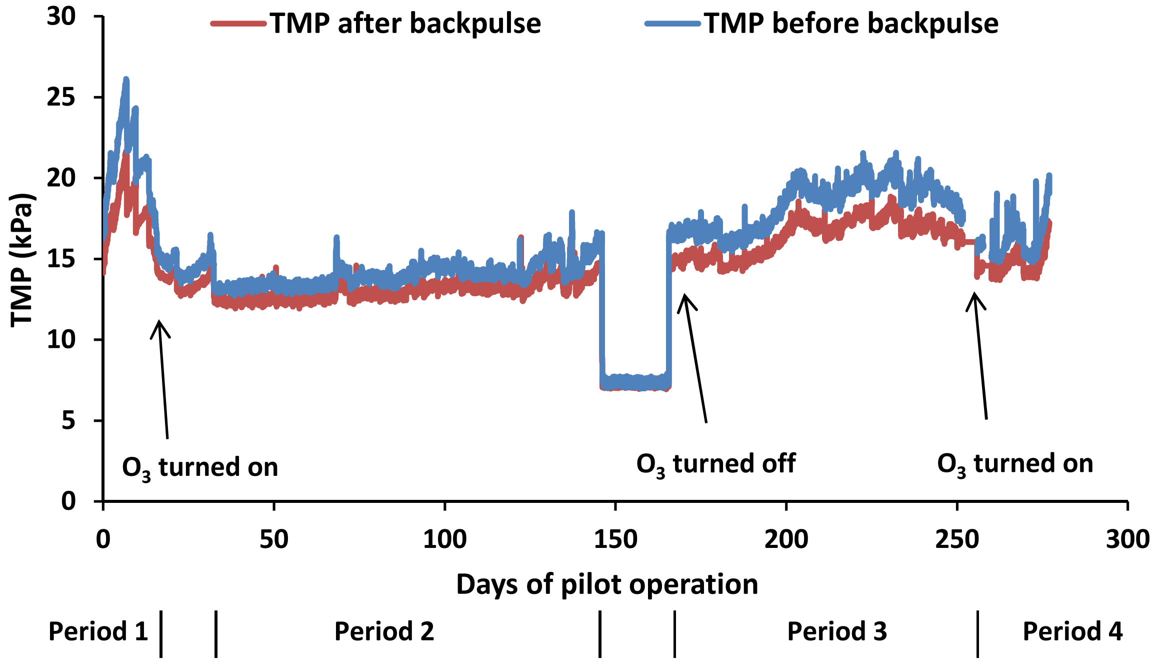

Figure 4.8 Pilot plant complete TMP data set corrected to 20°C before and after backpulse

In long-term membrane operation, the increase in TMP represents hydraulically irreversible fouling because hydraulically reversible fouling is controlled by membrane backpulse performed automatically every 30 minutes. The difference in TMP before and after backpulse, which is the amount of pressure reduced by backpulsing, represents hydraulically reversible fouling. Therefore, the slope of temperature corrected TMP data in Figure 4.7 is equal to the hydraulically irreversible fouling rate, and the difference between the two TMP data lines in Figure 4.8 can be used to determine hydraulically reversible fouling rate. There were two ozone online and two ozone offline periods during the complete period of pilot operation. Among those four periods, Period 1 (from day 1 to day 21) had the highest hydraulically reversible and irreversible fouling rates. This was likely due to the fact that the membrane module was new at the beginning of operation and the initial adsorption/deposition of hydrophobic foulants onto membrane surfaces is important at this stage (van Der Bruggen *et al.*, 2002). Period 4 (from day 250 on) had a very high hydraulically irreversible fouling rate and a relatively low hydraulically reversible fouling rate. This could be because testing occurred in a period when turbidity in both raw water and BACC#3 effluent were high (Figure 4.2).

Ozone was online in Period 2 and out of service in Period 3. Both periods had long-term stable operation and similar water quality in terms of turbidity and DOC concentration, which allowed for investigation into the impact of ozone on hydraulically reversible and irreversible fouling rates. TMP data before and after membrane backpulse during these two periods is presented in Figure 4.8 so that fouling rates can be determined and direct comparison between with and without ozone can be made. By comparing the slope and the difference between two TMP lines for these two periods, it is evident that both hydraulically reversible and irreversible fouling rates were higher in Period 3 with ozone out of service. In addition, hydraulically reversible fouling rates for Period 2 and Period 3 were calculated based on ΔTMP of each filtration cycle, and results are presented in Figure 4.9. Again, a major difference in hydraulically reversible fouling rates between the two periods can be seen. The overall average of hydraulically reversible fouling rate during ozone online was 1.9 kPa/h, and it increased to 3.7 kPa/h when ozone was out of service. In fact, hydraulically reversible fouling rates with ozone were approximately 50% lower than without ozone. The same effect was observed for hydraulically irreversible fouling.

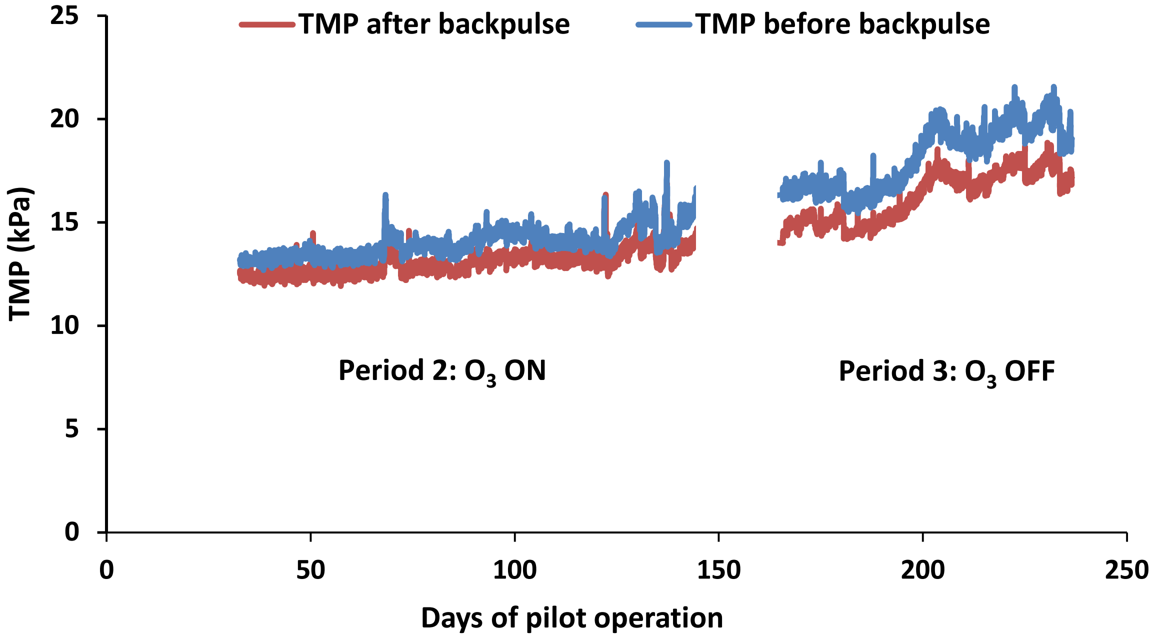

Figure 4.9 Selected TMP data corrected to 20°C before and after backpulse

Therefore, it can be concluded that ozonation pre-treatment was able to reduce both hydraulically reversible and irreversible fouling in downstream UF membranes. This finding is consistent with previous studies where ozonation was found to be able to reduce low pressure membrane fouling to various extents (Hashino *et al*., 2000 & 2001; Song *et al*., 2010; Geismar *et al*., 2012). For example, Geismar et al (2012) reported that at a transferred dosage of 1.0 mg/L, ozonation pre-treatment reduced UF membrane fouling by 44% as measured by the total unified membrane fouling index ($UMFI_T$). This finding is partially substantiated by the conclusion drawn previously that ozone was able to decrease biopolymer retention by UF membranes. Ozone decreased the average size of biopolymers, which have been identified as key foulants for the type of water and membrane investigated, resulting in less accumulation of foulants on membrane surfaces and within pores. Consequently, ozonation pre-treatment was able to reduce both hydraulically reversible and irreversible fouling in downstream UF membranes.

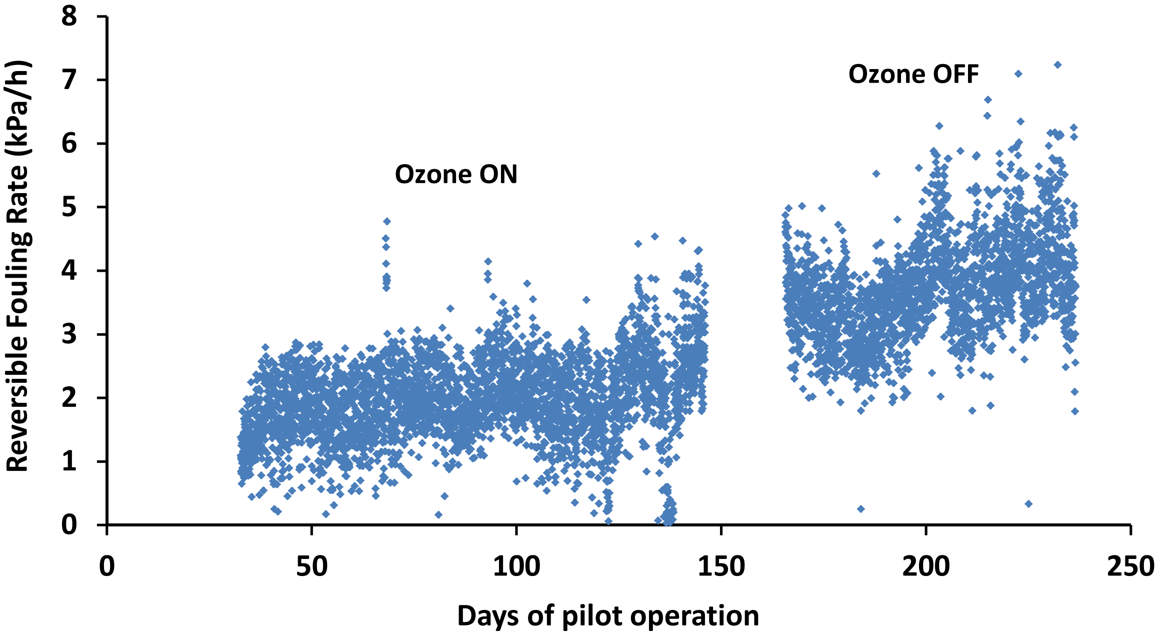

Figure 4.10 Hydraulically reversible fouling rate in each filtration cycle for Periods 2 and 3

4.4 Conclusions

Based on full-scale WTP monitoring and UF membrane pilot plant operation, the following conclusions can be drawn from this investigation into the impact of ozonation on the ozonation-biofiltration integrated pre-treatment process for UF membrane fouling control:

- Ozonation and biofiltration processes individually and in combination achieved good turbidity removal (despite the fact that water was not coagulated) but only minimal organic carbon removal.
- The operation of ozonation had clearly observable impacts on both biomass quantity and activity within the BACCs.
- Ozonation changed the hydrophobicity of DOC but not its concentration as it reduced specific UV absorbance (SUVA) by 43%.
- Among the different NOM fractions, biopolymers were the only fraction which was substantially removed by the UF membranes, implying that they were responsible for UF membrane fouling.
- Ozone decreased biopolymer retention in both full-scale and pilot-scale UF membranes
- With ozone online, hydraulically reversible and irreversible UF membrane fouling rates were 50% lower than when was ozone out of service.

4.5 Disclaimer

Mention of trade names or commercial products does not constitute endorsement or recommendation for their use by the authors or funding agencies.

Chapter 5 Ultrafiltration Membrane Performance with and without Ozonation-Biofiltration Pre-treatment

Summary

The integration of multiple pre-treatment processes is showing promise for fouling control in low pressure membrane (LPM) systems. Ozonation-biofiltration appears to be a new and promising process combination as a LPM pre-treatment. The purpose of this study was to assess the fouling reduction capacity by the integrated ozonation-biofiltration pre-treatment. In addition, the impact of foulants and water quality on the process was also investigated. The investigation was carried out at a full-scale water treatment plant which employs ozonation, biofiltration and ultrafiltration (UF) membranes. Three UF pilot plants were operated, one without pre-treatment, and the other two with ozonation-biofiltration as a pre-treatment. The performances of these pilot units were compared directly. The integrated ozonation–biofiltration pre-treatment was able to substantially reduce downstream UF membrane fouling, especially during times of elevated turbidity. In addition, elevated concentrations of biopolymers were found in pilot plant backpulse water, indicating that biopolymers are implicated in hydraulically reversible fouling. Hydraulically reversible fouling rate was found to be correlated to the concentration of biopolymers, and hydraulically irreversible fouling was largely determined by particle/colloid content.

5.1 Introduction

The use of low pressure membranes (LPMs) for drinking water treatment is increasing; however, fouling continues to be an important limitation to this technology as it can increase operation and maintenance costs and shorten membrane service life (Crittenden *et al.*, 2012).

LPM fouling mechanisms are quite complex, and are influenced by many factors, such as permeate flux, membrane type and feed water characteristics. Higher fouling rates have been demonstrated to occur more frequently at higher permeate flux (Field *et al*., 1995; Huang *et al*., 2007; Mosqueda-Jimenez & Huck, 2008). Membrane properties including pore size and surface charge can also significantly affect fouling behavior (AWWA, 2005; Amy, 2008). Feed water characteristics, in particular the content of natural organic matter (NOM) present in water is an important impacting factor for LPM fouling. Recent investigations have shown that organic fouling attributable to NOM is important for LPMs (Howe & Clark, 2002; Amy, 2008; Cai & Benjamin, 2011). It appears that the composition and size distribution of NOM is more important than the total concentration as only certain NOM components will contribute to LPM fouling. For example, Lee et al (2004) concluded that polysaccharides and proteins are important organic foulants for MF and UF membranes from among different NOM components. Haberkamp (2008) identified large humic substances and biopolymers, which consist of polysaccharides and protein-like substances, as the critical components of NOM responsible for organic fouling. Similarly, Hallé et al (2009) and Peldszus et al (2012) demonstrated the importance of biopolymers in both hydraulically reversible and irreversible fouling of LPMs. In addition, Peiris et al (2010) related high fouling events to elevated levels of particle/colloid-like material.

Membrane fouling can be mitigated by pre-treating the feed water. Effective pre-treatment can remove or alter significant portions of foulants before they reach the membranes, thereby reducing fouling. Potential LPM pre-treatment processes include adsorption, oxidation, coagulation, and filtration (Huang *et al*., 2009). Biological rapid filtration has more recently been identified as a promising pre-treatment option for LPMs. In addition to its ability to reduce microbial regrowth potential in distribution systems and to remove trace contaminants including those responsible for taste and odor (Rittmann, 1995; Urfer *et al*., 1997; Hozalski & Bouwer, 2001; Zhu *et al*., 2010), biofiltration has also been found to be able to reduce fouling in downstream LPMs (Park *et al*., 2002; Huck & Sozański, 2008; Hallé *et al*., 2009; Huck *et al*., 2011; Peldszus *et al*., 2011; Peldszus *et al*., 2012). Park et al (2002) used biofiltration followed by MF membranes to treat surface water and observed less fouling in a biofiltration-MF hybrid system than MF alone, and attributed this effect to the substantial turbidity and metal ion removal by the biofilter. Hallé et al (2009) and Peldszus et al (2012) further investigated fouling reduction mechanisms by biofiltration using hollow fibre polymeric UF membranes, and found that biofiltration was able to significantly reduce turbidity and biopolymer concentration in membrane influent, which if not removed, contributed to the fouling of UF membranes. The authors also investigated the impact of empty bed contact time (EBCT) and temperature on this process combination, and concluded that longer EBCTs and higher temperatures are beneficial for the performance of biofilters and reduction of membrane fouling. Although demonstrated to be a valid concept, biofiltration pre-treatment for LPMs still requires investigation at large-scale installations to further understand fouling control mechanisms, optimize operating parameters, and assess the potential of biological filtration in combination with other processes.

Ozonation is employed in drinking water treatment commonly for disinfection and oxidation of contaminants. Its applicability as an LPM pre-treatment has recently been recognized due to its ability to reduce membrane fouling by altering the composition and size distribution of dissolved organic matter (DOM) (Hyung *et al*., 2000; Wang *et al*., 2007). However, most studies investigating ozonation as an LPM pre-treatment have been in the context of wastewater and water reuse applications. Hashino et al (2000 & 2001), investigating ozone-resistant MF membranes following ozonation for drinking water treatment, found that the permeate flux was three to four times higher with pre-ozonation than without it.

An important development in membrane water treatment technology in recent years has been the integration of multiple pre-treatment processes. If membrane fouling can be effectively controlled by the integration of such pre-treatment combinations, the lifecycle costs of membrane systems may be reduced compensating for relatively higher initial capital costs. The process combination of ozonation and biofiltration (ozone prior to biological filtration) is a promising technology pairing. The ozonation-biofiltration-membrane (OBM) process is being employed in a large full-scale ultrafiltration drinking water treatment plant in the Region of Peel in Canada (Farr & Stampone 2007) and its use has been reported in a few studies. van der Hoek et al (2000) used ozone and biological activated carbon (BAC) filtration as pre-treatments for reverse osmosis (high pressure membranes), and experienced long-term stable operation without membrane cleaning. In an investigation using UF membranes to treat surface water, Osterhus et al (2007) concluded that ozone reactions are important in fouling control for OBM process. Geismar et al (2012) thoroughly examined the fouling control capacity of ozonation and BAC filtration as pre-treatments for different types of MF and UF membranes. The authors concluded that fouling in an OBM system is a function of ozone dosage and membrane type for a particular water, and somewhat unexpectedly found that biofiltration on its own did not contribute to significant fouling reduction. However, none of these studies was able to identify membrane foulants or investigate the impact of foulants on the OBM process because traditional water quality parameters used in these studies, such as TOC and DOC, cannot provide information on the composition and specific characteristics of NOM.

In the current study, a long-term detailed investigation into ozonation-biofiltration as an integrated LPM pre-treatment was undertaken in a large operating ultrafiltration drinking water treatment plant. This study focused on the (1) identification of membrane foulants, (2) assessment of fouling reduction capacity by the integrated pre-treatment process, (3) investigation of fouling control mechanisms, and (4) impact of water quality on the process.

5.2 Materials and Methods

5.2.1 Lakeview Water Treatment Plant

This study was conducted from January 2013 to May 2014 at the Lakeview Water Treatment Plant (WTP) owned by the Region of Peel located in Mississauga, Ontario, Canada, which draws water from Lake Ontario. The raw water characteristics after pre-chlorination for the study period were discussed in Chapter 3 and are summarized again in Table 5.1.

Table 5.1 Lakeview WTP raw water characteristics from January 2013 to May 2014 (n=58)

Parameters	Unit	Average	Minimum	Maximum
Temperature	°C	7.9	1.6	18.5
pH	-	7.61	7.08	7.99
Turbidity	NTU	0.47	0.24	1.64
Conductivity	µS/cm	318	255	403
Alkalinity	mg $CaCO_3$/L	89	75	102
TOC	mg C/L	2.06	1.61	2.69
DOC	mg C/L	2.01	1.59	2.60
SUVA	L/(mg C·m)	0.89	0.64	1.18

At the time of this research, the WTP had a capacity of 800 ML/d and consisted of a conventional treatment train and an advanced treatment train. The focus of this investigation was on the advanced treatment train of the plant. The advanced treatment train made up half of the plant capacity (i.e., 400 ML/d) and employed OBM processes (i.e., ozonation, biologically active carbon filtration, ultrafiltration membranes) as well as pre- and post-free chlorine application. The OBM2 facility was commissioned in July 2014 following the completion of this study. In OBM1 facility, where this study was conducted, there are 5 parallel biologically active carbon contactors (BACCs) and 12 membrane trains in operation. Ozone was out of service from January to March 2013, May to June 2013, and January to March 2014. When ozone was online, its dosage ranged from 1.5 to 2.2 mg/L assuming a pH of 8 and average summer (15°C) and winter (8°C) conditions, respectively (Region of Peel, 2011).

5.2.2 UF Pilot Plants

Three ZeeWeed®10 UF membrane pilot plants (GE Water and Process Technologies, Oakville, ON, Canada; Rocco Mazzaferro Services Inc., Waterdown, ON, Canada) were used in this study (Pilot#1, #2, and #3). Pilot#1 was operated from May 2013 to May 2014 and fed with the effluent from full-scale BACC#3. It contained a polyvinylidene fluoride (PVDF) hollow fibre ZeeWeed®500 membrane module with a nominal surface area of 1.0 m^2 and nominal pore size of 0.04 µm immersed in a membrane tank. Pilot#2 was operated from March to May 2014 with the Lakeview WTP's de-chlorinated raw water, and it contained the same type of membrane module as Pilot#1. De-chlorination was achieved by constantly dosing 0.1 mol/L sodium thiosulfate solution into the overhead feed tank of Pilot#2 using a peristaltic pump (Module 3385, Control Company, Friendswood, TX, USA). Pilot#3 was operated from March to May 2014 and was also fed with full-scale BACC#3 effluent; however, it was equipped with a ZeeWeed®1000 module with a nominal surface area of 1.0 m^2 and nominal pore size of 0.02 µm, which is the same type of membrane used at the full-scale Lakeview WTP.

The pilot units were operated in dead-end mode at a constant permeate flux of 52 L/m^2/h (LMH) with periodic backflushing. Water in the membrane tank was drawn by vacuum through fibre walls (outside-in) and ultimately into a permeate tank. A typical operation sequence included (1) permeation for 30 min, (2) back pulse with aeration for 30 s, (3) aeration for 15 s (4) draining of the tank with aeration, and (5) filling of the permeate tank for 2 min. Pilot#1 and Pilot#2 were programmed to continuously monitor and record data on temperature, flow, permeability, and trans-membrane pressure (TMP) with their built-in temperature sensors, flow meters, and pressure transducers. The purpose of Pilot#3 operation was to compare its permeate characteristics with those of Pilot #1. To examine the effect of reduced flux, Pilot#1 was operated at 26 LMH without human supervision and maintenance cleaning from December 17 2013 to January 6 2014. Except for the period mentioned, maintenance cleaning was performed manually twice a week (typically on Tuesdays and Fridays) by soaking the membrane in 100 mg/L sodium hypochlorite solution for 20 min. The membrane modules were all new at the start of operation. Membrane integrity tests and clean water permeability tests were conducted regularly to ensure sound membrane condition and effectiveness of cleaning. Fibre repairs were occasionally performed on Pilot#1 as required. Recovery cleanings were occasionally performed on Pilot#2 by soaking the membrane in 500 mg/L sodium hypochlorite solution for 8 - 10 h when its operating TMP reached 100 kPa.

5.2.3 Sample Collection and Analyses

Full-scale WTP monitoring was conducted biweekly from January to March 2013 and weekly from April 2013 to May 2014 (except for April 30, December 24, and 31 in 2013). Water samples were collected from the raw water intake (following screening and pre-chlorination), the effluents of the pre-ozonated (and occasionally non-pre-ozonated) biologically active carbon contactors 3 and 5 (BACC#3 and BACC#5), and in permeate from one of the full-scale membrane trains (UF#42) (Figure 5.1). Permeate from all three UF pilot plants was also collected during the operation of those units. In addition, six raw water samples prior to pre-chlorination were collected in February and March 2014; eight membrane backpulse samples were collected for Pilot#1 and #2 in May 2014.

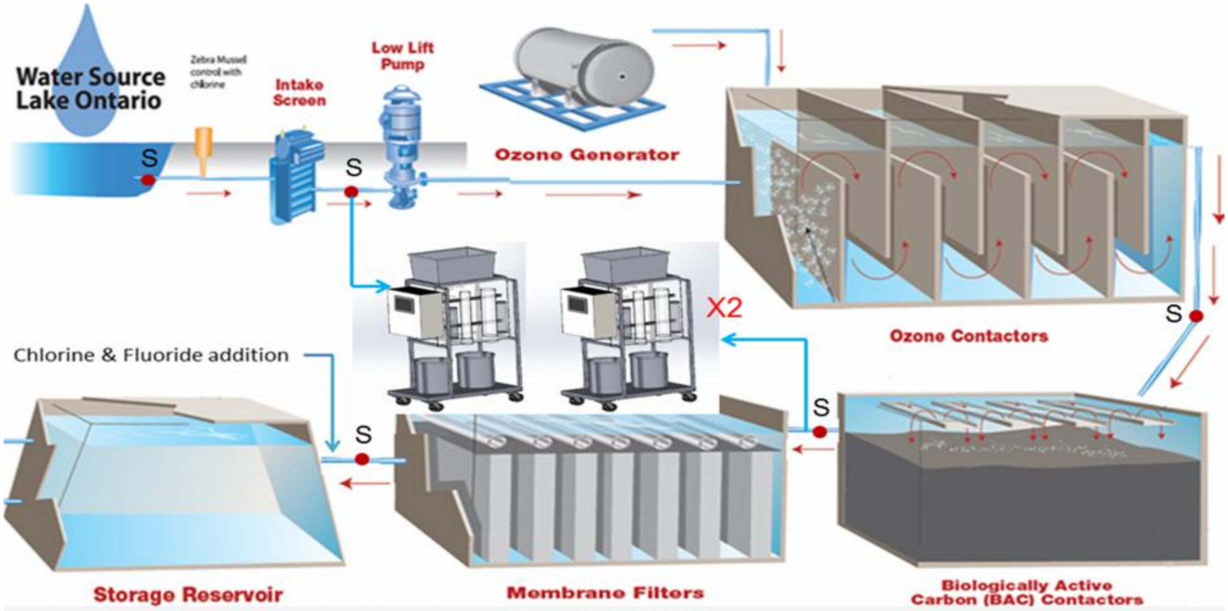

Figure 5.1 Lakeview WTP advanced treatment train process schematic with UF pilot plants and sampling locations. (S represents sampling locations) (Reproduced with permission from the Region of Peel)

The following water quality parameters were measured in some or all of the samples: pH, temperature, alkalinity, conductivity, turbidity, chlorine residual, UV_{254}, and specific UV absorbance (SUVA) using standard methods. Total and dissolved organic carbon (TOC/DOC) were measured using an OI-Analytical TOC analyzer model 1010 (College Station, TX, USA) from January to August 2013 and model 1030 from August 2013 to May 2014 by wet-oxidation method (Standard Methods 5310D). DOC samples were filtered through 0.45μm polyethersulfone filters and preserved with phosphoric acid to pH 2 before measurements. UV_{254} was measured with UV-Vis spectrometer (Cary 100, Agilent Technologies, Mississauga, ON) with no pH adjustment, and SUVA was calculated as follows: $SUVA = UV_{254}/DOC$. Liquid chromatography-organic carbon detection (LC-OCD) (DOC-Labor Dr. Huber, Karlsruhe, Germany) analysis was employed to measure the concentrations of the various NOM fractions including the biopolymers, humic substances, building blocks, low molecular weight (LMW) acids and humics, and LMW neutrals in water as described in Huber et al (2011). Total chlorine residual was measured onsite using a pocket colorimeter^{TM} II (Hach, Loveland, CO). Turbidity was monitored using online turbidimeters (Hach 1720E and FT660sc). Samples for the remaining parameters were analyzed at the University of Waterloo laboratories, and if not measured on-line, samples were stored at 4°C until analysis.

5.3 Results and Discussion

5.3.1 Pre-chlorination

At the Lakeview WTP, chlorine is added prior to the intake screen for zebra mussel control. The raw water characteristics before and after pre-chlorination based on 6 samples collected in February and March 2014 are presented in Table 5.2. Paired t-tests ($\alpha = 0.05$) indicated that pre-chlorination slightly reduced raw water alkalinity but there were no significant differences between pre- and post-chlorination water samples with respect to parameters investigated in this study. In general, the source water at this plant is of high quality.

The concentrations of NOM fractions in raw water pre- and post-chlorination are shown in Figure 5.2. The LMW acids and LMW humics concentrations were so low as to be negligible. Student t-tests ($\alpha = 0.05$) confirmed that pre-chlorination did not significantly change the concentration of any of the NOM fractions measured by LC-OCD. Overall, it can be concluded that pre-chlorination did not significantly alter the characteristics of the raw water except in the case of alkalinity. While the chlorine may oxidize some compounds present in the water, the applied chlorine dosages do not appear to be high enough to substantially redistribute the concentrations of the NOM groups being measured in this study.

Table 5.2 Lakeview WTP pre- and post-chlorination raw water characteristics in February and March 2014 (n=6)

Parameters	Unit	Before pre-chlorination	After pre-chlorination
pH	-	7.50	7.33
Conductivity	µS/cm	320	321
Alkalinity	mg $CaCO_3$/L	96	90
TOC	mg C/L	1.68	1.73
DOC	mg C/L	1.63	1.66
SUVA	L/(mg C·m)	0.85	0.92

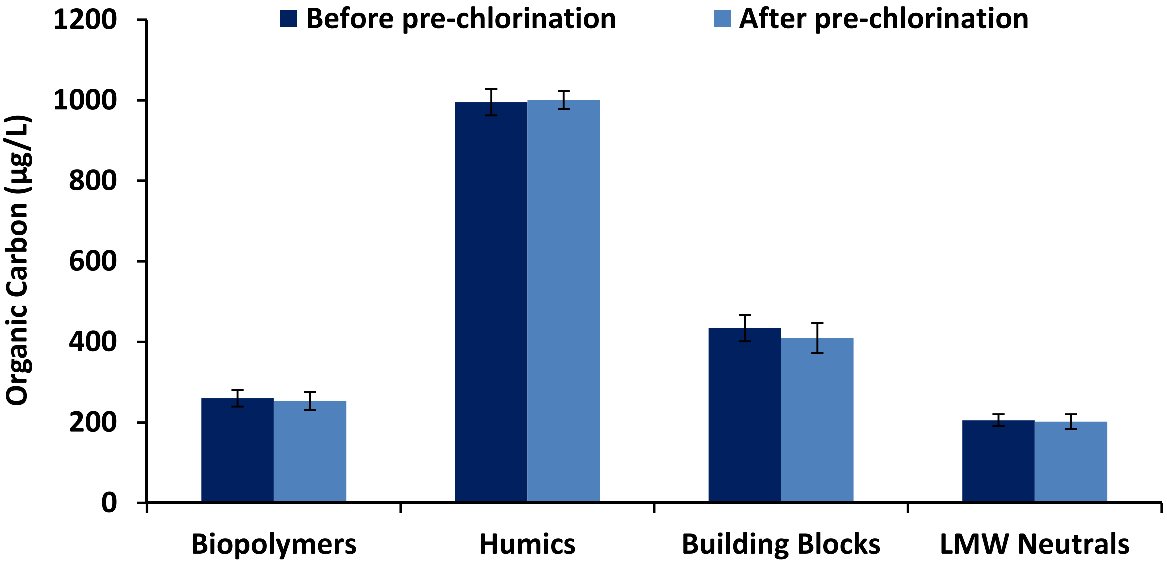

Figure 5.2 Concentrations of different NOM fractions before and after pre-chlorination. Error bars show standard deviation.

5.3.2 Flux and Membrane Fouling

UF Pilot#1 was operated at a permeate flux of 26 L/m^2/h (LMH) without maintenance cleaning from December 17 2013 to January 6 2014 (day 146 to day 165) and at 52 LMH with twice weekly maintenance cleaning for the remainder of the test period. From day 35 to day 250, the pilot unit was in stable operation without rapid changes in feed water (BACC#3 effluent) quality or membrane fouling rate. This period included pilot operation at both fluxes as well as periods of ozone being online and out of service (The effect of ozone on membrane fouling rate has been previously discussed in Chapter 4). The trans-membrane pressure (TMP) data corrected to 20°C captured by Pilot#1 before and after membrane backpulses during this period are illustrated in Figure 5.3. In long-term membrane operation, the increase in TMP represents hydraulically irreversible fouling because the hydraulically reversible fouling was controlled by membrane backpulse performed automatically every 30 minutes. The difference in TMP before and after backpulse, which is the amount of pressure reduced by backpulsing, represents the hydraulically reversible fouling. Therefore, the slope of TMP data line in Figure 5.3 represents the hydraulically irreversible fouling rate, and the difference between the two TMP data lines can be used to determine hydraulically reversible fouling rate. The numerical calculation of TMP temperature correction and hydraulically reversible and irreversible fouling rates have been discussed in Chapter 4.

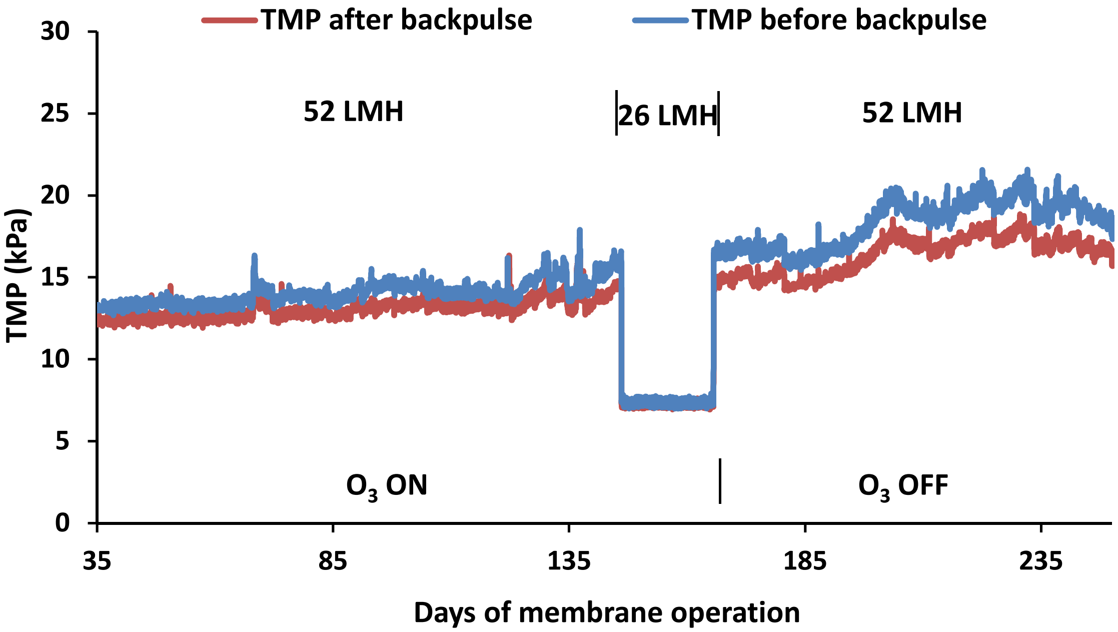

Figure 5.3 Pilot#1 selected TMP data corrected to 20°C at two fluxes of 26 and 52 LMH

From Figure 5.3, it is evident that both hydraulically reversible and irreversible fouling rates were significantly lower during low permeate flux pilot operation. In fact, the hydraulically irreversible fouling rate in the Pilot#1 membrane at 26 LMH was so low that it could essentially be neglected for the roughly 20 d that the pilot plant was run at this condition. The average hydraulically reversible fouling rate at 26 LMH was 0.4 kPa/h. In contrast, at 52 LMH, the average hydraulically reversible fouling rate was 1.9 kPa/h with ozone online and 3.7 kPa/h with ozone offline (4.75 and 9.25 times greater). These results are consistent with previous membrane fouling studies in which researchers concluded that higher fouling rates generally occur at higher permeate flux for membranes operating in constant flux mode and the extent of fouling increase with increasing permeate flux is a function of both membrane type and feed water characteristics (Field *et al*., 1995; Huang *et al*., 2007; Mosqueda-Jimenez & Huck, 2008).

5.3.3 Performance of Different Membranes Types

Two types of membranes at two different scales were used in this study. Pilot#3 contained the same type of membrane module as the full-scale membranes employed at the Lakeview WTP. Pilots#1 and #2 contained a membrane module with a larger fibre diameter. Except for the low flux test period on Pilot#1, all three pilot units were operated under the same conditions and at the same permeate flux as a single full-scale membrane train (UF#42) to allow for direct comparison. The performances of these different types of membranes with respect to DOC removal and biopolymer retention are presented in Table 5.3. As shown in the table, the performances of the two types of pilot-scale membranes with respect to DOC and biopolymer removal were very similar. The full-scale and pilot-scale membranes also performed similarly except that the full-scale membranes had a slightly higher biopolymer retention rate. Therefore, it can be concluded that under the same operating conditions, data collected from pilot-scale and full-scale membranes were comparable.

Table 5.3 Comparison of different types of membranes used in the study

Membrane	Fibre	Flux (LMH)	DOC Removal (%)	Biopolymer Retention (%)	n
UF Full-scale	ZeeWeed® 1000	52	12 (±6)	64 (±6)	58
UF Pilot#1 & #2	ZeeWeed® 500	52	13 (±5)	59 (±7)	50
UF Pilot#3	ZeeWeed® 1000	52	11 (±2)	59 (±11)	7

n = number of samples

5.3.4 Biopolymers and Membrane Fouling

During this investigation, the DOC in incoming raw water at the Lakeview WTP consisted of 13.3% biopolymers, 49.6% humic substances, 18.8% building blocks, 10.4% low molecular weight (LMW) neutrals, and less than 0.1% LMW acids and humics as per measured by LC-OCD (by weight). Concentrations of the target NOM fractions at different treatment stages including Pilot#1 and Pilot#2 permeate from LC-OCD measurements are presented in Figure 5.4 to provide insights into the transformation of the different NOM fractions through the treatment process train and the identification of UF membrane foulants.

Among all NOM fractions measured by LC-OCD, biopolymers were the only fraction that underwent a significant change through the entire treatment process train and were substantially retained by both full-scale and pilot-scale UF membranes. Retention here is defined as the net removal of the fraction being investigated through an individual process. Specifically, 61%, 56%, and 54% of biopolymers in the feed water were retained by the full-scale, Pilot#1, and Pilot#2 membranes, respectively. Virtually no removal of humics and other fractions was observed. This strongly implies that biopolymers are the fraction of NOM responsible for UF membrane fouling. This finding is consistent with recent studies where biopolymers or polysaccharides and protein-like substances were found to be key foulants for drinking water LPMs (Her *et al*., 2007; Hallé *et al*., 2009; Peldszus *et al*., 2012; Rahman 2014) and membrane bioreactors (Jiang *et al*., 2010).

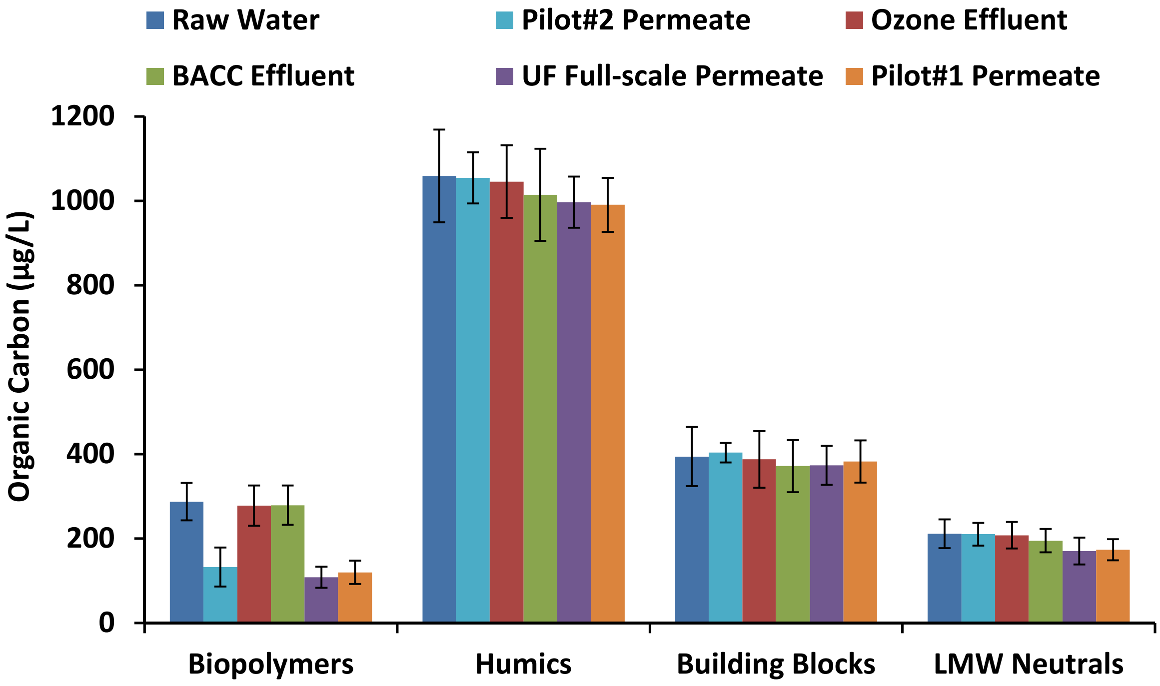

Figure 5.4 Concentrations of different NOM fractions following each treatment stage including pilot permeate. Pilot#1 was fed with BACC effluent, and Pilot#2 was fed with raw water. Error bars show standard deviation (n = 33 for Pilot#1 permeate, n = 10 for Pilot#1 permeate, and n = 38 for other samples).

To confirm that biopolymers did in fact contribute to fouling, backpulse water samples were collected from the two pilot units and analyzed by LC-OCD. Concentrations of the different NOM fractions in the feed, permeate, and backpulse water samples from Pilot#1 and Pilot#2, based on four LC-OCD measurements, are presented in Figures 5.5 and 5.6, respectively. By comparing the two figures, it can be seen that the concentrations of humic substances, building blocks, and LMW neutrals were relatively consistent in feed, permeate, and backpulse water for both pilot-scale membranes. In contrast, concentrations of biopolymers were significantly lower in permeate samples and slightly higher in backpulse samples than feed samples for both pilots. This observation further suggests that biopolymers are retained by UF membranes during filtration and removed during backpulsing, linking biopolymers to hydraulically reversible fouling. A precise evaluation of the actual amount of material deposited on membrane during filtration would require a mass balance calculation; however, this observation can provide evidence of the rejection of biopolymers by UF membranes. The accumulation of biopolymers appears to occur on the surfaces of UF membranes due to their large molecular weights, contributing to cake layer formation. This result cannot provide insight on hydraulically irreversible fouling because the concentrations of biopolymers remaining on membrane surface and within pores could not be individually measured.

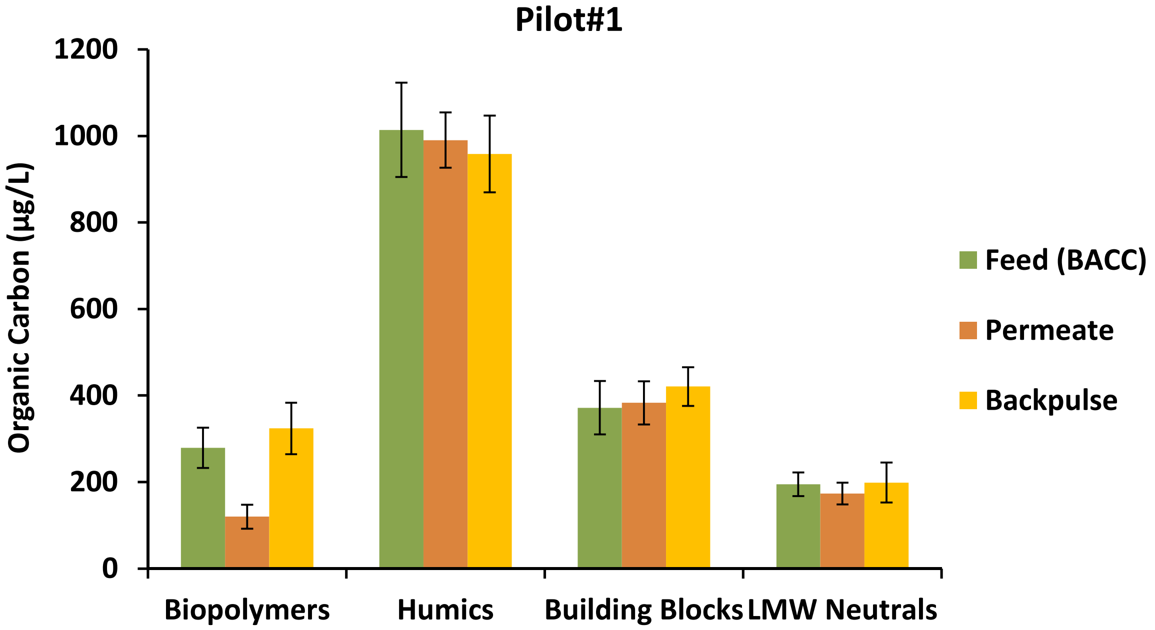

Figure 5.5 Concentrations of NOM fractions in the feed, permeate, and backpulse samples from Pilot#1 fed with BACC#3 effluent (n = 4). Error bars show standard deviation.

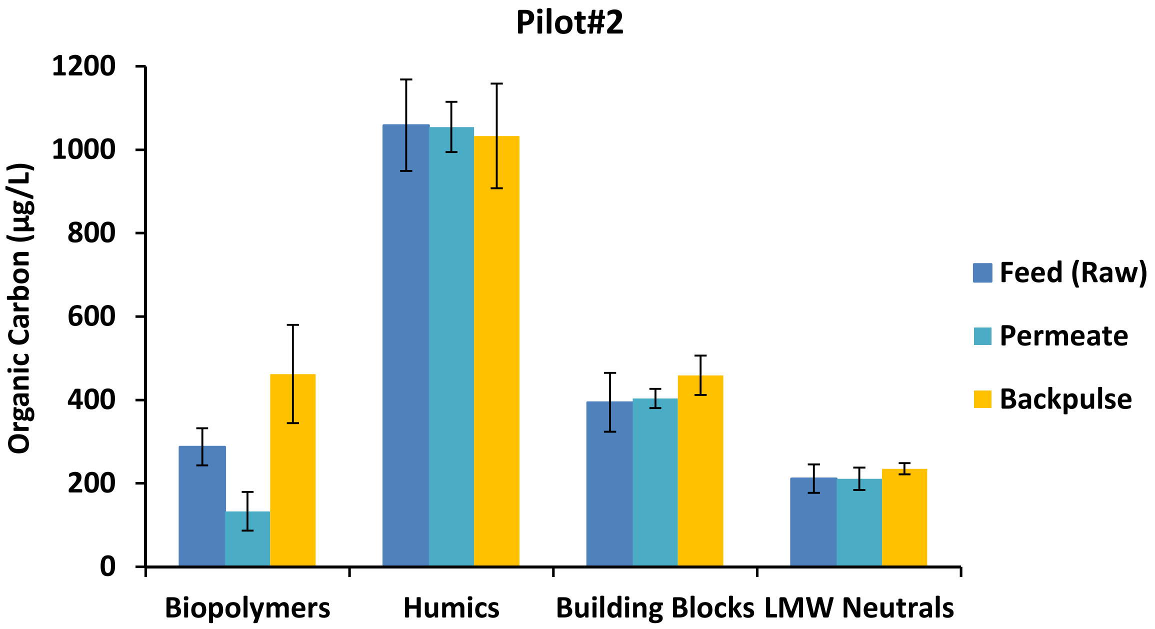

Figure 5.6 Concentrations of NOM fractions in the feed, permeate and backpulse sample of Pilot#2 fed with raw water (n = 4). Error bars show standard deviation.

Since biopolymers are retained (rejected) by UF membranes and are very likely to contribute to fouling, their concentration in membrane feed water is an important factor to consider. The relationship between the concentration of biopolymers in feed water and hydraulically reversible and irreversible fouling rates for Pilot#1 with ozone online are presented in Figure 5.7 and 5.8. Hydraulically reversible fouling rates were calculated using the average ΔTMP (change in TMP during a filtration cycle) from five filtration cycles including the cycle when a water sample was collected for LC-OCD analysis, and two cycles before and after. Hydraulically irreversible fouling rates were calculated using TMP in the last filtration cycle before two consecutive maintenance cleanings. From the two figures, it can be seen that for Pilot#1, the hydraulically reversible fouling rate was linearly correlated to biopolymer concentration in the feed water (which was BACC#3 effluent); whereas the hydraulically irreversible fouling rate was not related to biopolymer concentration. Hallé et al (2009) reported similar findings in bench-scale UF membranes of the same type treating a different water source.

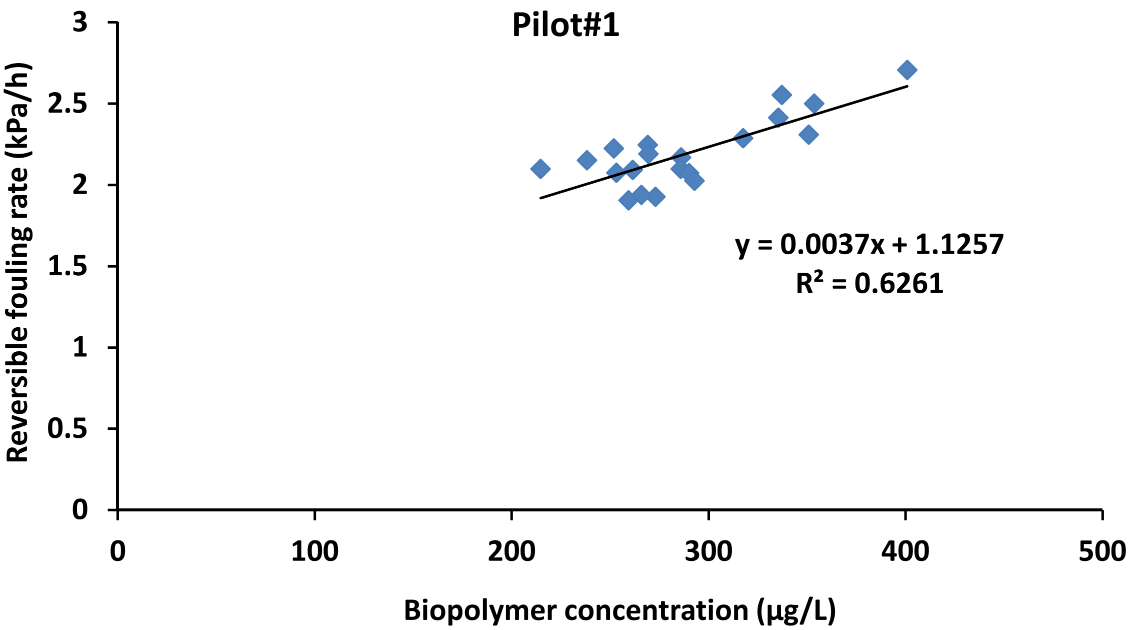

Figure 5.7 Pilot#1 feed water biopolymer concentration vs. irreversible fouling rate

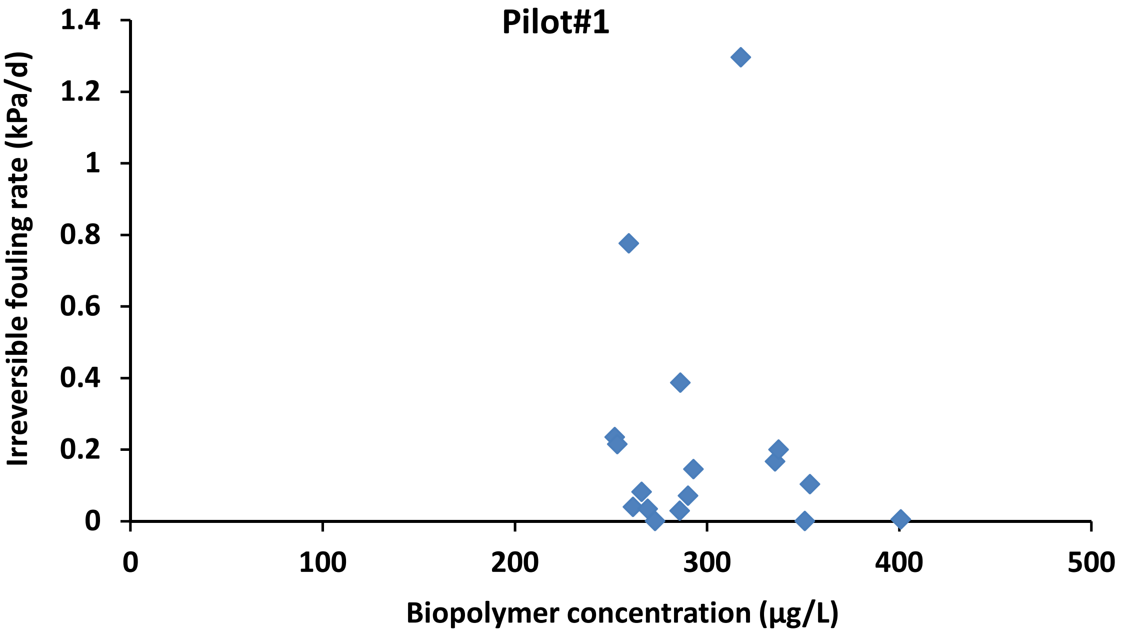

Figure 5.8 Pilot#1 feed water biopolymer concentration vs. irreversible fouling rate

Biopolymer concentrations in feed water (raw water) vs. hydraulically reversible and irreversible fouling rates for Pilot#2 are presented in Figures 5.9 and 5.10. As was the case for Pilot#1, the biopolymer concentration in the Pilot#2 feed water was in the 200 - 400 µg/L range, and the hydraulically irreversible fouling was not related to the concentration of biopolymers. However, unlike Pilot#1, which was fed with BACC effluent, the hydraulically reversible fouling rate for Pilot#2, which was fed with raw water, was not correlated to biopolymer concentration. This appears to be due to the fact that the hydraulically reversible fouling rate for Pilot#2 was so low that it could not be reliably detected (mostly below 0.5 kPa/h). Therefore, no correlation between hydraulically reversible fouling and biopolymer concentration could be established for Pilot#2. Overall, biopolymer concentration was correlated to hydraulically reversible fouling rate. The contribution of biopolymers to hydraulically irreversible fouling in the raw water was either minimal or obscured by particle/colloid content of this water. Biopolymers composition rather than concentration likely plays an important role in hydraulically irreversible fouling (Hallé *et al*., 2009).

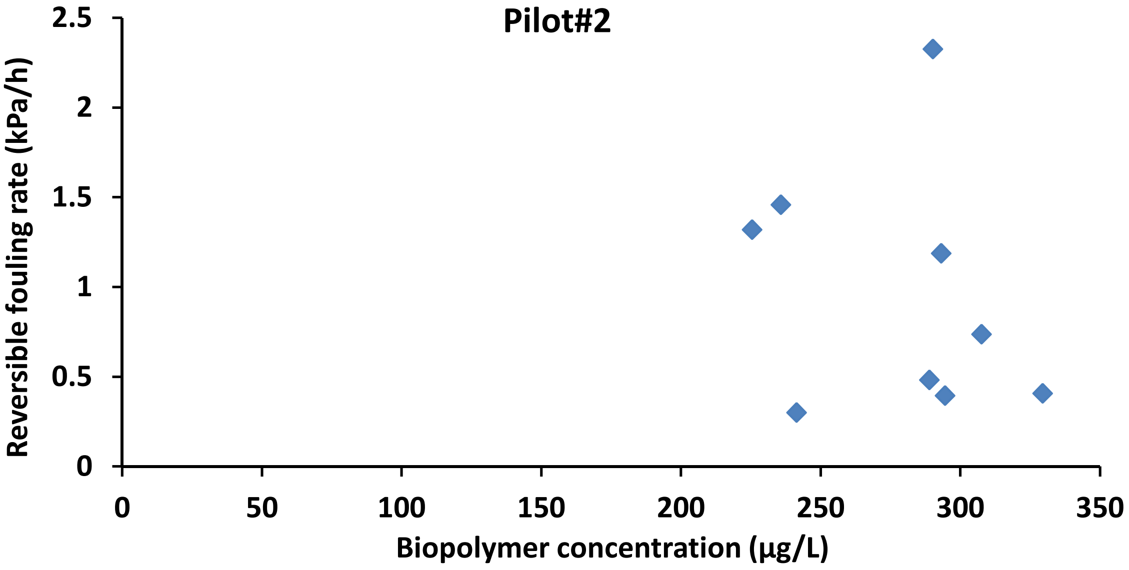

Figure 5.9 Pilot#2 feed water biopolymer concentration vs. reversible fouling rate

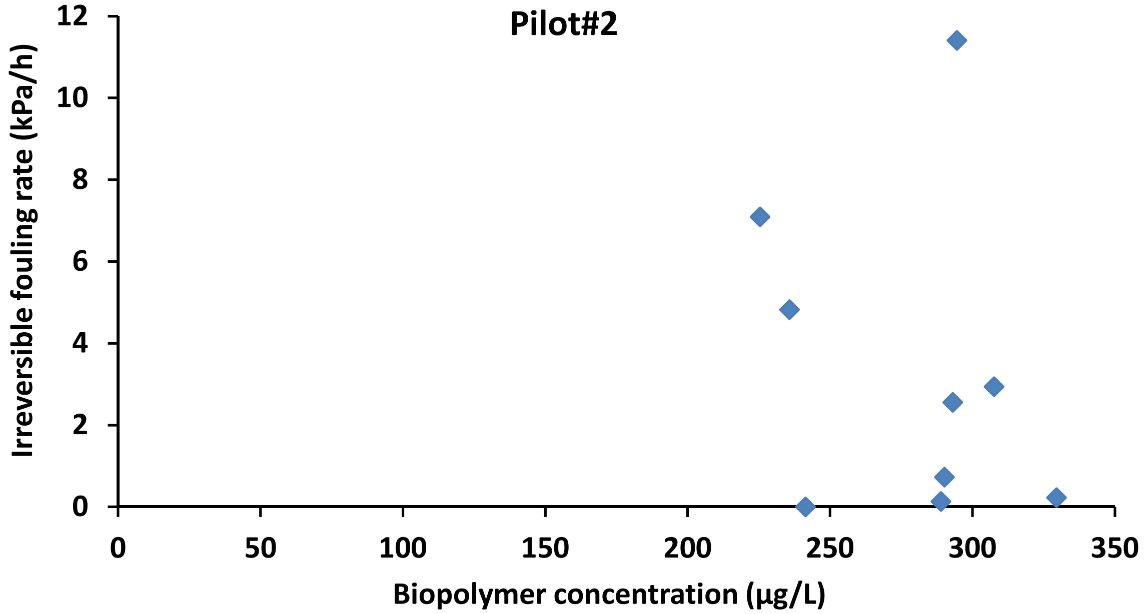

Figure 5.10 Pilot#2 feed water biopolymer concentration vs. irreversible fouling rate

During this investigation, the biopolymer concentration in the Lakeview WTP incoming raw water ranged from 0.20 to 0.38 mg/L. The average biopolymer concentration in the raw water was 0.28 mg/L, accounting for approximately 13% of the DOC. Raw water biopolymer and DOC concentrations from January 2013 to May 2014 are presented in Figure 5.11. Unlike the particle/colloid content, both the DOC and biopolymer concentrations in raw water were relatively consistent throughout the investigation period. In addition, the proportion of biopolymers in DOC was also consistent ranging from 11% to 15%. Higher DOC and biopolymer concentrations appeared more frequently at higher temperatures, but this seasonal phenomenon was less apparent than in previous studies conducted in Southern Ontario (Pharand, 2014; Rahman *et al.*, 2014). This is very likely because those studies were conducted on river waters with wide temperature ranges, whereas this investigation was carried out with water from a very large lake which is much less influenced by air temperature and organic/inorganic inputs.

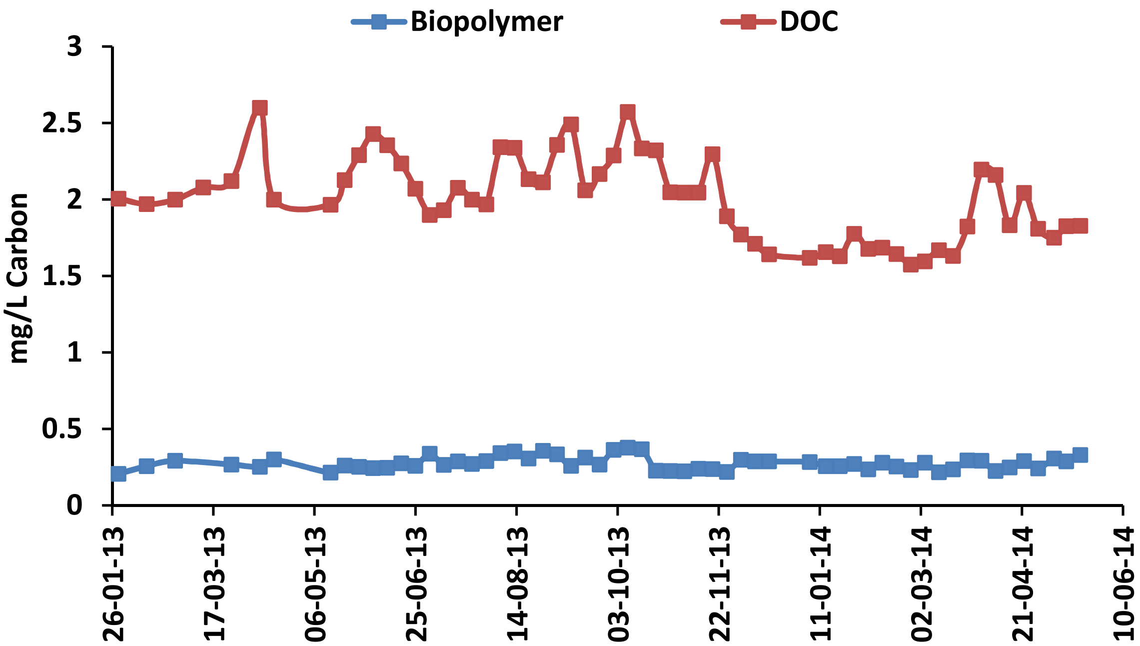

Figure 5.11 Raw water DOC and biopolymer concentration from January 2013 to May 2014 (n = 58)

5.3.5 Membrane Performance with and without Ozonation-Biofiltration Pre-treatment

Pilot#1 and Pilot#2 were operated under the same conditions from March to May 2014 for approximately two months. Ozonation had been out of service since January 2014 and continued to be offline during first month of the test period. It then was brought back online for the second month of the test period. Pilot#1 was fed with effluent from full-scale BACC#3; therefore it had biofiltration only as a pre-treatment in the first month and ozonation-biofiltration in the second month of the test period. Pilot#2 was fed with de-chlorinated plant raw water, so it did not have any pre-treatment. Trans-membrane pressure (TMP) data after each backpulse corrected to 20°C from the two pilot units during test period are presented in Figure 5.12.

The difference between TMP immediately before and after backpulse can be used to determine hydraulically reversible fouling rate. In the first 30 d can be seen that Pilot#1 had higher hydraulically reversible fouling rates than Pilot#2 whose TMPs (before and after backpulse) are essentially overlapping. This observation is also consistent with the hydraulically reversible fouling rates of Pilot#1 and Pilot#2 calculated numerically and presented in Figure 5.7 and 5.9. The difference in hydraulically reversible fouling rates is likely because the membrane in Pilot#2 was new at the beginning of the test period, whereas Pilot#1 had been in operation for 10 months prior to the test period. A cake layer would likely have already formed during this time, which contributed to higher hydraulically reversible fouling rate in Pilot#1. As can be seen, the hydraulically reversible fouling rate decreased in Pilot#1 when ozone came back into service (at around day 35), indicating that ozonation was likely able to control cake layer formation. The impact of ozone on membrane fouling has been discussed in detail in Chapter 4.

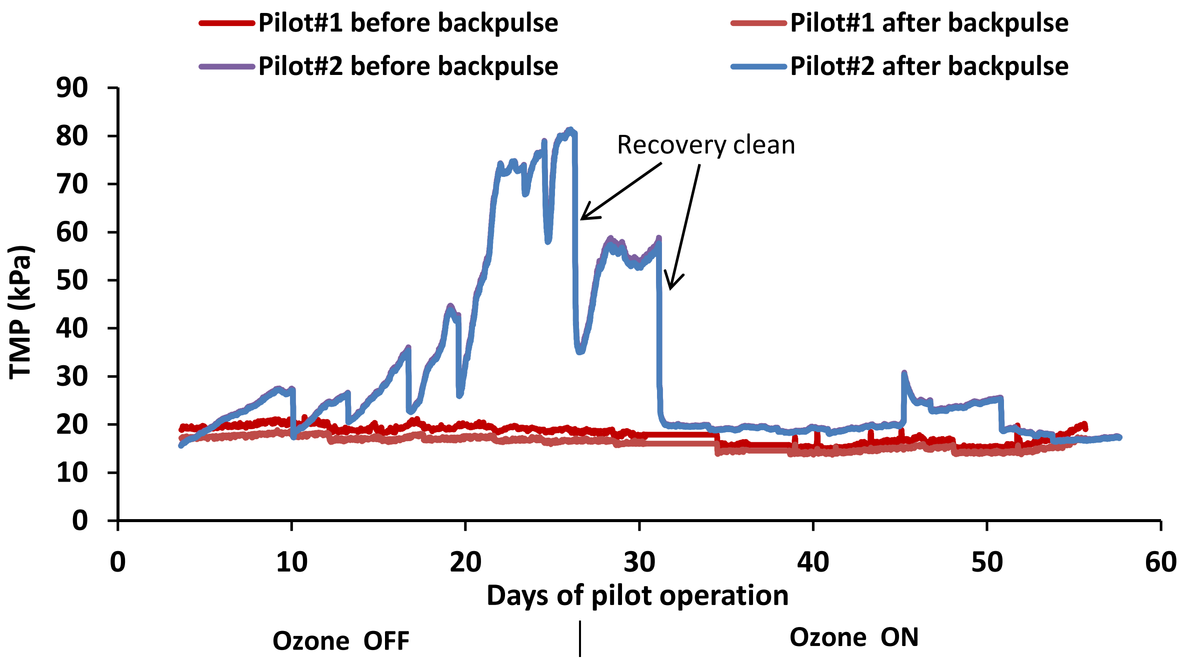

Figure 5.12 Pilot#1 and #2 TMP data before and after each backpulse (corrected to 20°C)

The slopes of the TMP trend lines in Figure 5.12 represent the hydraulically irreversible fouling rates. Pilot#2 (fed with raw water) had substantially higher hydraulically irreversible fouling rates than Pilot#1 from day 1 to day 32 and from day 45 to day 50. In fact, the hydraulically irreversible fouling rates were so high that recovery cleaning had to be performed twice on Pilot#2. This confirms that the biofiltration and the tandem ozonation-biofiltration pre-treatments were able to substantially reduce hydraulically irreversible fouling in the downstream UF membranes. Hydraulically irreversible fouling rates in Pilot#2 which did not have any pre-treatment varied substantially during the test period. This was likely a result of changes in its feed water (raw water) characteristics. However, since the DOC and biopolymer concentrations in the raw water remained relatively consistent (varied within 30%) during the test period, the higher variation in hydraulically irreversible fouling rates appears to have been caused by changes in the particle/colloid content of the raw water.

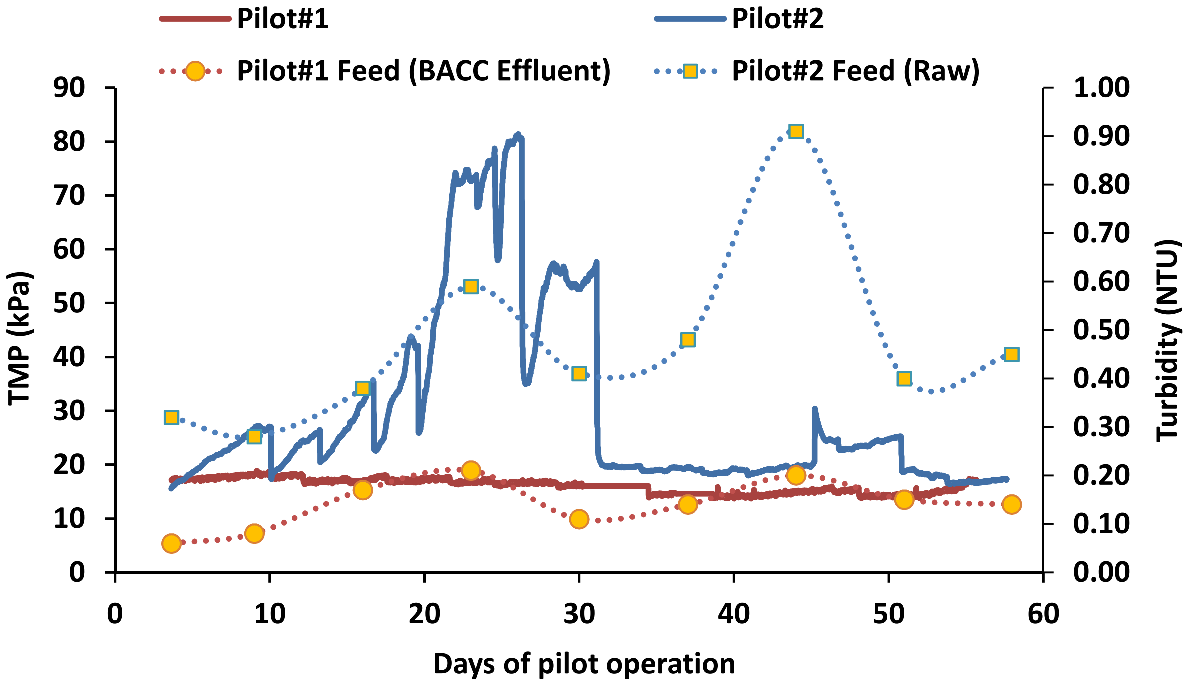

Figure 5.13 Pilot#1 and #2 TMP after each backpulse (corrected to 20°C) vs. feed water turbidity data

Figure 5.13 illustrates the relationship between feed water turbidity and hydraulically irreversible fouling rate expressed by TMP after backpulse from the two pilot units. The feed water turbidity for Pilot#1 was substantially lower than Pilot#2. In fact, the integrated ozonation-biofiltration pre-treatment process was able to remove 58 – 88% of incoming turbidity, which directly resulted in lower hydraulically irreversible fouling rates in Pilot#1. Also, it can be observed from the figure that the hydraulically irreversible fouling rate was correlated to feed water turbidity for Pilot#2. Hydraulically irreversible fouling rate increased with increasing turbidity, and hydraulically irreversible fouling rate increase dramatically at two turbidity peaks. Therefore, it can be concluded that hydraulically irreversible fouling of UF membranes is related to the particle/colloid content in feed water. This finding is in line with the conclusion drawn earlier that the contribution of biopolymers to hydraulically irreversible fouling rate was insignificant. Similar to present study, Howe & Clark (2002) concluded that fouling in LPM systems was predominantly attributable to small organic colloids. Peiris et al (2010) related high fouling events to elevated levels of particulate/colloidal-like material. In addition, Mosqueda-Jimenez & Huck (2006a) analyzed the foulant layer for the same type of membrane as used in this study, and found that the material in the foulant layer was mostly in particulate form.

5.4 Conclusions

By analyzing NOM fractions at different treatment process stages and comparing membrane performance in the pilot units, the following conclusions can be drawn from this investigation into hydraulically reversible and irreversible fouling in LPMs and the control of fouling by the integrated ozonation-biofiltration pre-treatment process:

- Pre-chlorination did not significantly alter the incoming raw water characteristics of interest at the Lakeview WTP.
- Both hydraulically reversible and irreversible fouling rates were significantly lower at reduced permeate flux.
- The performance of pilot-scale and full-scale membranes was similar with respect to DOC and biopolymer removal.
- Elevated concentrations of biopolymers were found in pilot plant backpulse water, indicating that biopolymers are implicated in UF membrane fouling, especially hydraulically reversible fouling.
- Hydraulically reversible fouling rates were correlated to biopolymer concentration, while hydraulically irreversible fouling was largely determined by particle/colloid content (for this water).
- The integrated ozonation–biofiltration pre-treatment process can remove particles and colloids thereby substantially reducing hydraulically irreversible fouling, especially during times of elevated turbidity.

5.5 Disclaimer

Mention of trade names or commercial products does not constitute endorsement or recommendation for their use by the authors or funding agencies.

Chapter 6 Conclusions and Recommendations

The research presented in this thesis was conducted to: 1) investigate the effect of ozonation prior to biofiltration as a low pressure membrane (LPM) pre-treatment, 2) assess the fouling reduction capacity of integrated ozonation-biofiltration pre-treatment, and 3) investigate the impact of natural organic matter (NOM) and water quality on the process. The ultimate goal was to evaluate integrated ozonation-biofiltration LPM pre-treatment using natural water at a large scale and provide useful considerations for the design and operation of membrane water treatment facilities. The Lakeview Water Treatment Plant (WTP) provided a unique opportunity for this research as it is one of the few WTPs in the world which employs an ozonation, biofiltration, and ultrafiltration process sequence. Long-term monitoring of the plant water quality at different treatment stages allowed for investigation into the impact of changes in membrane feed water quality on fouling. In addition, the operation of UF pilot plants provided an opportunity to compare UF membrane performance with and without pre-treatment under controlled operating conditions so that the impact of fouling reduction capacity by the pre-treatment processes and ozone can be determined. Liquid chromatography-organic carbon detection (LC-OCD) analysis provided valuable information on the transformation of NOM through the ozonation-biofiltration-membrane (OBM) process train and allowed for the identification of organic foulants.

6.1 Integration of Findings and Conclusions

Water quality monitoring was conducted at the Lakeview WTP for 16 months using traditional parameters such as TOC/DOC and SUVA as well as a novel LC-OCD method to characterize NOM at different stages in the plant’s OBM treatment train. In addition, the biomass quantity and activity within the biologically active carbon contactors (BACCs) were analyzed. Three UF pilot plants were used in this study to investigate membrane fouling. Pilot#1 was operated with BACC effluent for one year which covered two ozone-on and two ozone-off periods. This allowed for investigation into the effect of ozone on fouling control by the pre-treatment processes. Pilot#1 was also operated at two different permeate fluxes which provided insights into the relationship between flux and fouling rate. Pilot#2 was operated with the plant's incoming raw water for two months under the same operating conditions as Pilot#1 to provide direct comparison and assess the fouling control capacity of the ozonation-biofiltration pre-treatment process. Pilot#3 was used to compare the performance of different types of membranes including full-scale and pilot-scale membranes. Under the conditions investigated, the following conclusions can be made:

- The incoming raw water of the Lakeview WTP had low levels of organic carbon, relatively low temperature, and occasional small turbidity peaks. The seasonal variations in these parameters were small. In addition, pre-chlorination did not significantly alter the monitored raw water characteristics.
- The full-scale plant ozonation and biofiltration processes individually, and in combination, achieved good turbidity removal but only minimal organic carbon removal. Specifically, the ozonation-biofiltration process combination achieved 2 – 10% DOC removal and 58 – 88% turbidity removal.
- Good correlation was found between ATP and FDA which are indicators of biomass quantity and activity. The operation of ozonation clearly impacted on both biomass quantity and activity within the BACCs. Biomass quantity and activity within the BACCs increased substantially when ozone came back into service. Ozonation was

able to reduce the hydrophobicity of DOC as suggested by a 43% reduction of specific UV absorbance (SUVA), thus making the DOC more biodegradable.

- The pilot- and full-scale UF membranes consistently achieved 90% turbidity removal and 12% DOC removal. Compounds in that 12% of the DOC were likely membrane foulants.
- Biopolymers made up 13% of DOC in the water investigated, and the concentration of biopolymers in raw water was relatively consistent. Approximately 60% of biopolymers were removed by both full-scale and pilot-scale UF membranes implying that they were responsible for fouling. This suggests that less than 8% of the total DOC in raw water accounted for fouling at this plant. No other NOM fractions measured by LC-OCD were significantly removed by the UF membranes.
- Elevated concentrations of biopolymers were found in the pilot UF membrane backpulse water, which further indicated that biopolymers can contribute to UF membrane fouling, especially hydraulically reversible fouling. No other NOM fraction was detected at increased concentrations in the backpulse water.
- The hydraulically reversible fouling rate was found to be correlated to the concentration of biopolymers, and hydraulically irreversible fouling was largely determined by the presence of particles and colloids.
- Ozonation enhanced fouling control in downstream UF membranes. With ozone online, hydraulically reversible and irreversible UF membrane fouling rates were 50% lower than when ozone out of service. In addition, ozone decreased biopolymer retention in both full-scale and pilot-scale membranes which supported the finding that ozonation can reduce membrane fouling.

- Both hydraulically reversible and irreversible fouling rates were significantly lower at reduced permeate fluxes.
- The integrated ozonation–biofiltration pre-treatment process can remove particles and colloids thereby substantially reducing hydraulically irreversible fouling, especially during elevated turbidity events.

6.2 Implications for Membrane Water Treatment

Based on the results of this study, the following recommendations are made for the design and operation of membrane water treatment facilities:

- Ultrafiltration is a robust process which is consistently able to produce high quality and microbiologically safe finished water. Therefore, more consideration should be given to UF membranes when designing municipal water treatment plants. Ultrafiltration water treatment is a sustainable and forward-looking strategy to cope with challenges including limited plant and carbon footprints, stricter regulations, and unpredictable source water quality variations in the future.
- Membrane fouling can be controlled with appropriate pre-treatment(s). The integrated ozonation-biofiltration pre-treatment process can effectively reduce fouling in downstream UF membranes, especially hydraulically irreversible fouling; thus the plant operating cost can be decreased and membrane service life can be extended. The example of the Lakeview WTP's OBM process train can be copied by other utilities, especially those with large populations.
- Ozonation is important in fouling control in the OBM process. Ozone should be operated continuously to ensure optimum membrane performance. For utilities where

ozonation is installed to control seasonal taste and odor issues like the Lakeview WTP, decisions should be made based on total energy efficiency including energy required by ozone generation and energy saved from reduced membrane fouling.

- Biopolymers, particles, and colloids are major fouling contributors. Their concentrations, as well as seasonal variations in source water are important parameters for process selection and design of membrane water treatment facilities.
- Biofilters should be designed and operated to maximize biopolymer removal for better fouling control, especially in the case of hydraulically reversible fouling.
- Operating flux is very important as it relates to membrane fouling. Reduced flux can substantially reduce the fouling rate for any plant. This should be taken into account during design taking both capital cost and operating conditions into consideration.
- Pilot-scale investigations can provide useful information on proposed processes and are recommended during the design phase of membrane water treatment facilities.

6.3 Recommendations for Future Research

This study has provided some insights into low pressure membrane fouling and pre-treatment and also raised some questions. The following recommendations are suggested for future research:

- Previous studies have found that biofiltration can substantially remove biopolymers. However, the biofilters investigated in this study achieved only minimal biopolymer removal under conditions which were expected to lead to better removals. Further research should investigate the relationship between biopolymer removal and biofilter operating parameters as well as the water characteristics. Focus should be on the optimization of biofilter operation and biopolymer removal.

- Coagulation is known to be a successful LPM pre-treatment. Research should be conducted to compare the fouling control capacities of ozonation-biofiltration to coagulation.
- Membrane fouling is influenced by both water characteristics and membrane type. Different membrane types, such as ceramic membranes and different water sources, particularly those with higher biopolymer concentrations and greater seasonal variations should be tested to assess the range of effectiveness of ozonation-biofiltration as a LPM pre-treatment.
- This study revealed that biopolymers and particles/colloids are major contributors to hydraulically reversible and irreversible fouling, respectively. Also observed was the potential of ozone to decrease biopolymer retention in downstream membranes (without biofiltration). Future research should be performed at the nano-level to investigate the accumulation mechanisms of these materials on membrane surfaces and within pores; and on ozone reaction pathways with biopolymers.

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Appendix A: Photos and Additional Information of UF Pilot Plants

Figure A-1. UF pilot#1 front view showing

The primary components of the system are:

1. ZW-1000 Jr. module; total membrane area: ~1 m^2
2. Membrane tank
3. Micropump metering pump, maximum 1.2 L/m @ 0.3 – 1.2 bar TDH, reversible

4. Air pump for membrane scouring
5. Electric actuated 2-way and 3-way control valves
6. Backpulse tank, 9 liter capacity, with overflow
7. Chemical wash tank, 9 liter capacity, with overflow
8. Control panel with PLC and operator interface.
9. Pressure transmitter
10. Flow Transmitter

Figure A-2. UF pilot#1 back view

Figure A-3. UF Pilot#3 side view showing mechanical and electrical components

Figure A-4. UF Piot#3 side view showing ZeeWeed500® membrane module tank (left), ZeeWeed1000® membrane module tank (middle), and chemical cleaning tank (right)

Pilot#2 and #3 were exactly the same. Both units contained two membrane tanks designated for two types of membrane modules. Pilot#2 was operated with a ZeeWeed500® membrane module; Pilot#3 was operated with a ZeeWeed1000® membrane module.

Table A-1. ZeeWeed500® and ZeeWeed1000® membrane module properties (Information extracted from material fact sheets provided by GE Water & Process Technologies)

Parameters	ZeeWeed500®	ZeeWeed1000®
Material	PVDF	PVDF
Nominal pore size	0.04 µm	0.02 µm
Surface properties	Non-ionic & hydrophilic	Non-ionic & hydrophilic
Fibre diameter	1.9 mm OD/ 0.8 mm ID	0.95 mm OD/ 0.47 mm ID
Flow path	Outside-in	Outside-in
Operating TMP range	-90 to 90 kPa	-90 to 90 kPa
Max operating temperature	40°C	40°C
Operating pH range	5.0 – 9.5	5.0 – 10.0
Cleaning pH range	2.0 – 10.5	2.0 – 12.0 @ ≤30°C 2.0 – 11.5.0 @ 31 - 40°C
Max Cl_2 concentration	1,000 ppm	1,000 ppm pH ≤ 10.5 500 ppm pH ≤ 11.5

Figure A-5. UF pilot#1 (right) and #3 (left) in parallel operation

Figure A-6. Pilot#2 ZeeWeed500® membrane module at the beginning of operation

Figure A-7. Fouled Pilot#2 ZeeWeed500$^{®}$ membrane module during operation before recovery cleaning

Appendix B: Additional Lakeview Water Treatment Plant Operational Data

Table B-1. Lakeview Water Treatment Plant Operational Data

Date	Cl_2 dosage (mg/L)	O_3 dosage (mg O_3/mg C)	BACC#3 flow rate (MLD)	UF#42 flow rate (MLD)	UF#42 TMP (kPa)	Plant flow demand (MLD)
Jan-29-13	1.40	0	36.72	22.35	-42	200
Feb-12-13	1.60	0	28.88	22.41	-41	150
Feb-26-13	1.55	0	0	0	-	-
Mar-26-13	1.55	0	70.00	0	-	220
Apr-09-13	1.55	0.80	40.75	35.75	-42	160
Apr-16-13	1.60	1.00	64.50	35.67	-66	234
May-14-13	1.70	0	76.04	35.65	-57	238
May-21-13	1.50	0	62.15	35.70	-58	240
May-28-13	1.50	0	63.71	35.50	-59	260
Jun-04-13	1.60	0	58.58	35.73	-58	240
Jun-11-13	1.70	1.10	36.20	35.52	-49	165
Jun-18-13	1.60	0.90	33.49	35.68	-46	150
Jun-25-13	1.55	0.80	59.65	35.57	-51	310
Jul-02-13	1.60	1.10	36.90	35.69	-38	200
Jul-09-13	1.60	0.85	64.84	35.65	-52	310
Jul-16-13	1.60	1.00	88.22	0	-	350
Jul-23-13	1.60	0.90	68.74	35.57	-51	350
Jul-30-13	1.55	0.80	34.00	35.62	-51	160
Aug-06-13	1.60	1.10	51.25	35.56	-49	240
Aug-13-13	1.55	1.00	53.42	35.65	-48	240
Aug-20-13	1.45	0.80	69.14	35.72	-55	350
Aug-27-13	1.55	1.00	46.79	18.94	-20	200
Sep-03-13	1.60	0.75	42.77	35.57	-42	200
Sep-10-13	1.45	0.80	64.95	0	-	310
Sep-17-13	1.40	1.10	47.44	35.41	-49	240
Sep-24-13	1.50	1.10	37.48	35.63	-49	190
Oct-01-13	1.60	0.90	53.08	35.60	-43	200
Oct-08-13	1.35	1.10	74.18	35.61	-43	270
Oct-15-13	1.45	1.25	48.66	35.71	-41	200

Table B-1. Lakeview Water Treatment Plant Operational Data (con't)

Date	Cl_2 dosage (mg/L)	O_3 dosage (mg O_3/mg C)	BACC#3 flow rate (MLD)	UF#42 flow rate (MLD)	UF#42 TMP (kPa)	Plant flow demand (MLD)
Oct-22-13	1.42	0.70	29.30	35.72	-60	150
Oct-29-13	1.27	0.60	42.12	35.70	-63	200
Nov-05-13	1.40	0.85	46.00	34.90	-57	160
Nov-12-13	1.40	0.75	58.19	35.68	-68	260
Nov-19-13	1.40	0.65	51.13	36.17	-65	240
Nov-26-13	1.40	0.80	47.68	35.80	-60	160
Dec-03-13	1.40	0.75	71.89	35.36	-76	270
Dec-10-13	1.40	0.85	69.41	35.33	-81	240
Dec-17-13	1.30	0.90	60.87	0	-	250
Jan-06-14	1.40	0	0	34.23	-81	220
Jan-14-14	0	0	0	0	-	-
Jan-21-14	1.40	0	0	0	-	-
Jan-28-14	1.40	0	33.91	35.36	-79	140
Feb-04-14	0	0	0	0	-	-
Feb-11-14	1.35	0	38.83	36.22	-79	150
Feb-18-14	1.45	0	31.14	33.92	-83	150
Feb-25-14	1.36	0	35.68	34.8	-81	150
Mar-04-14	1.35	0	33.03	35.09	-79	140
Mar-11-14	1.37	0	40.14	34.10	-82	130
Mar-18-14	1.40	0	35.59	34.64	-81	120
Mar-25-14	1.37	0	32.42	34.21	-81	140
Apr-01-14	1.58	1.05	42.09	33.62	-79	160
Apr-08-14	1.38	1.00	54.06	34.07	-81	200
Apr-15-14	1.40	0.95	62.03	0	-	160
Apr-22-14	0.8	1.00	56.87	35.54	-74	230
Apr-29-14	1.30	0.70	69.05	35.56	-68	260
May-07-14	1.40	1.00	57.39	35.53	-64	230
May-13-14	1.30	0.85	70.73	35.63	-71	310
May-20-14	1.35	0.85	54.15	35.41	-73	220

*Trans-membrane pressure (TMP) is driven by vacuum; therefore it has negative value.

Appendix C: Biomass Quantity (ATP) and Activity (FDA) Data

Table C-1. BACC#3 raw ATP and FDA data

Data	ATP (ng ATP/cm^3)	FDA (µg FDA/cm^3)
Feb-26-13	21.7	-
Mar-12-13	14.3	-
Mar-26-13	13.9	-
Apr-16-13	41.4	-
May-14-13	21.7	62.8
May-28-13	28.1	54.9
Jun-11-13	59.6	105.6
Jun-25-13	186.3	106.1
Jul-09-13	78.1	76.4
Jul-23-13	222.0	113.1
Aug-06-13	182.2	113.4
Aug-20-13	556.3	123.4
Sep-03-13	184.9	253.9
Sep-17-13	426.4	137.2
Oct-01-13	550.8	186.3
Oct-15-13	490.0	79.6
Oct-29-13	483.8	157.9
Nov-12-13	187.0	98.7
Nov-26-13	211.6	132.3
Dec-10-13	244.2	145.2
Jan-06-14	274.8	213.4
Jan-21-14	169.9	78.1
Feb-04-14	20.8	40.8
Feb-18-14	33.4	55.3
Mar-04-14	25.4	54.0
Mar-18-14	23.7	36.4
Apr-01-14	22.7	43.2
Apr-15-14	27.1	35.9
Apr-29-14	88.3	45.8
May-13-14	95.4	89.1

Appendix D: Water Quality Raw Data

Table D-1. Raw water quality data

Date	Temperature (°C)	pH	Conductivity (µS/cm)	Turbidity (NTU)	Alkalinity (mg/L $CaCO_3$)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
Jan-29-13	3.63	7.91	255	0.35	102	2.10	1.97	0.018	0.91
Feb-12-13	3.64	7.96	269	0.43	98	2.05	2.00	0.019	0.95
Feb-26-13	3.32	7.93	325	0.49	96	2.13	2.08	0.018	0.84
Mar-26-13	4.08	7.63	310	0.51	96	2.69	2.60	0.022	0.87
Apr-09-13	6.35	7.56	290	0.66	97	2.10	2.00	0.018	0.90
Apr-16-13	6.03	7.54	317	1.47	88	2.29	1.97	0.023	1.15
May-14-13	7.48	7.68	280	0.28	85	2.12	2.13	0.016	0.76
May-21-13	9.98	7.89	282	0.36	82	2.12	2.29	0.018	0.77
May-28-13	10.03	7.89	271	0.34	84	2.58	2.43	0.019	0.80
Jun-04-13	9.67	7.69	290	0.38	85	2.25	2.36	0.018	0.77
Jun-11-13	12.46	7.83	305	0.54	85	2.18	2.24	0.019	0.86
Jun-18-13	13.11	7.88	293	0.39	84	2.04	2.07	0.019	0.92
Jun-25-13	11.52	7.67	295	0.31	82	1.56	1.90	0.017	0.91
Jul-02-13	18.53	7.31	304	0.62	84	2.22	1.93	0.017	0.89
Jul-09-13	11.78	7.36	305	0.37	83	2.03	2.08	0.019	0.92
Jul-16-13	14.55	7.44	315	0.86	87	2.00	2.00	0.023	1.17
Jul-23-13	10.06	7.36	310	0.45	87	2.03	1.97	0.022	1.09
Jul-30-13	11.86	7.51	312	0.38	86	2.28	2.34	0.021	0.89
Aug-06-13	12.77	7.54	309	0.40	86	2.31	2.34	0.019	0.83
Aug-13-13	12.72	7.55	310	0.43	85	2.26	2.13	0.021	0.99
Aug-20-13	11.61	7.51	317	0.31	84	2.12	2.11	0.021	1.00
Aug-27-13	16.86	7.08	310	0.61	84	2.37	2.36	0.021	0.88
Sep-03-13	16.67	7.36	310	0.58	84	2.26	2.49	0.019	0.76
Sep-10-13	9.88	7.44	308	0.33	78	2.49	2.06	0.020	0.96
Sep-17-13	13.57	7.69	308	0.36	84	2.21	2.17	0.019	0.87
Sep-24-13	12.28	7.49	312	0.42	86	2.20	2.29	0.018	0.77
Oct-01-13	17.17	7.99	310	0.50	75	2.56	2.57	0.009	0.34

Table D-1. Raw water quality data (con’t)

Date	Temperature (°C)	pH	Conductivity (µS/cm)	Turbidity (NTU)	Alkalinity (mg/L $CaCO_3$)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
Oct-08-13	17.37	7.95	308	0.58	83	2.37	2.34	0.018	0.77
Oct-15-13	17.27	7.98	306	0.43	93	2.36	2.32	0.019	0.83
Oct-22-13	7.20	7.22	313	0.26	84	2.05	2.05	0.017	0.84
Oct-29-13	6.87	7.35	314	0.25	81	2.03	2.04	0.015	0.74
Nov-05-13	6.85	7.48	312	0.47	85	2.03	2.05	0.015	0.74
Nov-12-13	7.00	7.51	313	0.24	85	2.31	2.30	0.019	0.81
Nov-19-13	6.75	7.35	314	0.26	83	1.88	1.89	0.017	0.87
Nov-26-13	5.76	7.54	312	0.26	84	1.84	1.77	0.015	0.83
Dec-03-13	4.64	7.51	327	0.34	89	1.69	1.71	0.017	1.00
Dec-10-13	4.21	7.48	324	0.33	87	1.71	1.64	0.015	0.91
Dec-17-13	2.84	7.39	328	1.64	88	1.65	1.62	0.015	0.91
Jan-06-14	1.63	7.60	357	1.63	90	1.68	1.66	0.020	1.18
Jan-14-14	3.69	7.63	342	0.25	90	1.63	1.63	0.015	0.95
Jan-21-14	2.73	7.63	370	0.25	95	1.81	1.78	0.011	0.64
Jan-28-14	2.73	7.67	309	0.24	88	1.69	1.68	0.014	0.84
Feb-04-14	3.40	7.69	312	0.24	90	1.66	1.69	0.015	0.89
Feb-11-14	3.02	7.63	315	0.25	86	1.67	1.64	0.016	0.95
Feb-18-14	2.48	7.63	326	1.07	88	1.59	1.58	0.015	0.98
Feb-25-14	2.77	7.61	316	0.26	91	1.62	1.60	0.016	1.01
Mar-04-14	2.49	7.69	324	0.25	91	1.64	1.67	0.015	0.88
Mar-11-14	2.79	7.69	349	0.27	95	1.66	1.63	0.013	0.82
Mar-18-14	2.48	7.56	333	0.32	94	2.18	1.82	0.016	0.86
Mar-25-14	2.81	7.68	367	0.28	92	2.43	2.20	0.014	0.64
Apr-01-14	3.50	7.56	403	0.38	95	2.45	2.16	0.018	0.83
Apr-08-14	3.95	7.49	349	0.59	95	2.12	1.83	0.015	0.84
Apr-15-14	5.23	7.57	350	0.41	96	2.30	2.04	0.016	0.77
Apr-22-14	5.97	7.61	358	0.48	96	1.83	1.81	0.019	1.06
Apr-29-14	5.86	7.56	338	0.91	94	1.76	1.75	0.015	0.86
May-07-14	7.45	7.69	368	0.40	98	1.89	1.83	0.024	1.30
May-13-14	8.45	7.68	347	0.45	96	1.85	1.83	0.018	0.98
May-20-14	7.07	7.65	337	0.32	97	1.78	1.76	0.016	0.90

Table D-2. Ozone effluent water quality data

Date	Temperature (°C)	pH	Conductivity (µS/cm)	Turbidity (NTU)	TOC (mg/L)	DOC (mg/L)	UV$_{254}$ (cm^{-1})	SUVA (L/(mg C·m))
Jan-29-13	11.0	7.50	266	-	2.10	2.11	0.020	0.95
Feb-12-13	8.5	7.57	263	-	2.05	2.00	0.019	0.95
Feb-26-13	10.6	7.55	337	-	2.13	2.08	0.026	1.21
Mar-26-13	10.8	7.56	290	-	2.57	2.45	0.022	0.87
Apr-09-13	10.1	7.70	291	-	2.10	2.00	0.023	0.90
Apr-16-13	9.2	7.46	317	-	2.08	2.00	0.025	1.21
May-14-13	12.1	7.72	282	-	2.19	2.19	0.017	0.79
May-21-13	13.0	7.80	278	-	2.34	2.19	0.020	0.91
May-28-13	12.9	7.85	271	-	2.38	2.23	0.017	0.75
Jun-04-13	13.0	7.65	291	-	2.29	2.24	0.019	0.84
Jun-11-13	14.5	7.73	311	-	2.08	1.99	0.011	0.50
Jun-18-13	14.6	7.81	297	-	2.01	2.05	0.013	0.63
Jun-25-13	18.6	7.53	299	-	1.80	1.51	0.012	0.79
Jul-02-13	20.2	7.71	308	-	2.00	1.94	0.010	0.50
Jul-09-13	16.0	7.29	307	-	2.04	1.96	0.012	0.59
Jul-16-13	18.0	7.52	315	-	2.00	2.00	0.018	0.90
Jul-23-13	15.6	7.34	311	-	1.94	1.92	0.013	0.69
Jul-30-13	15.7	7.42	312	-	2.30	2.22	0.014	0.61
Aug-06-13	18.7	7.57	312	-	2.30	2.24	0.011	0.49
Aug-13-13	16.2	7.38	311	0.15	2.06	2.08	0.013	0.64
Aug-20-13	14.6	7.43	315	0.19	2.07	2.05	0.014	0.66
Aug-27-13	17.9	7.46	312	0.20	2.30	2.22	0.013	0.59
Sep-03-13	20.3	7.28	312	0.17	2.33	2.20	0.010	0.47
Sep-10-13	13.6	7.29	312	0.08	2.00	2.04	0.012	0.61
Sep-17-13	14.8	7.30	312	0.10	2.15	2.13	0.010	0.47
Sep-24-13	12.8	7.25	312	0.35	2.11	2.11	0.008	0.39
Oct-01-13	17.4	7.33	310	0.25	2.36	2.30	0.009	0.34
Oct-08-13	17.6	7.69	308	0.49	2.27	2.25	0.007	0.30
Oct-15-13	17.0	7.65	307	0.24	2.36	2.28	0.005	0.24
Oct-22-13	8.7	7.26	313	0.09	2.01	2.09	0.008	0.35

Table D-2. Ozone effluent water quality data (con't)

Date	Temperature (°C)	pH	Conductivity (µS/cm)	Turbidity (NTU)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
Oct-29-13	8.6	7.47	312	0.11	2.00	1.96	0.011	0.58
Nov-05-13	8.9	7.51	311	0.09	2.00	1.95	0.007	0.34
Nov-12-13	7.7	7.43	310	0.14	2.31	2.26	0.008	0.33
Nov-19-13	8.5	7.48	313	0.08	1.90	1.95	0.011	0.55
Nov-26-13	6.4	7.63	310	0.09	1.75	1.73	0.059	0.34
Dec-03-13	5.5	7.25	327	0.18	1.68	1.68	0.010	0.57
Dec-10-13	5.4	7.36	324	0.10	1.62	1.62	0.007	0.46
Dec-17-13	4.0	7.17	326	1.48	1.61	1.61	0.009	0.56
Jan-06-14	3.5	7.29	352	0.35	1.68	1.64	0.019	1.14
Jan-14-14	3.5	7.48	331	0.35	1.69	1.66	0.016	0.95
Jan-21-14	3.8	7.18	368	0.19	1.81	1.79	0.011	0.64
Jan-28-14	5.4	7.48	312	0.10	1.67	1.66	0.015	0.87
Feb-04-14	6.1	7.55	317	0.11	1.65	1.65	0.013	0.76
Feb-11-14	5.4	7.28	315	0.12	1.67	1.63	0.015	0.90
Feb-18-14	4.2	7.39	327	0.76	1.69	1.56	0.015	0.98
Feb-25-14	4.8	7.22	313	0.12	1.59	1.57	0.015	0.96
Mar-04-14	5.8	7.47	315	0.11	1.76	1.72	0.016	0.90
Mar-11-14	6.2	7.77	346	0.15	1.67	1.63	0.014	0.83
Mar-18-14	5.6	7.42	330	0.24	1.93	1.87	0.015	0.77
Mar-25-14	5.8	7.38	387	0.17	2.31	2.27	0.014	0.62
Apr-01-14	6.2	7.36	402	0.34	2.29	2.07	0.018	0.88
Apr-08-14	7.0	7.24	351	0.43	2.03	2.08	0.015	0.73
Apr-15-14	8.6	7.22	353	0.22	2.02	2.08	0.009	0.43
Apr-22-14	10.2	7.34	361	0.22	1.75	1.73	0.011	0.64
Apr-29-14	10.1	7.22	350	0.31	1.73	1.72	0.009	0.52
May-07-14	10.7	7.44	372	0.20	1.92	1.79	0.015	0.85
May-13-14	11.7	7.20	345	0.13	1.81	1.80	0.011	0.59
May-20-14	11.7	7.39	340	0.10	1.70	1.66	0.007	0.44

Table D-3. BACC#3 effluent water quality data

Date	Temperature (°C)	pH	Conductivity (µS/cm)	Turbidity (NTU)	TOC (mg/L)	DOC (mg/L)	UV$_{254}$ (cm^{-1})	SUVA (L/(mg C·m))
Jan-29-13	4.0	7.50	281	0.12	2.14	2.13	0.023	1.08
Feb-12-13	3.8	7.57	259	0.18	2.13	2.00	0.019	0.95
Feb-26-13	3.3	7.55	328	0.32	2.15	2.05	0.019	0.92
Mar-26-13	4.3	7.56	299	0.25	2.57	2.44	0.015	0.99
Apr-09-13	6.4	7.70	288	0.30	2.10	2.00	0.017	0.85
Apr-16-13	6.7	7.46	324	0.70	2.01	1.91	0.020	0.99
May-14-13	8.2	7.72	282	0.09	2.15	2.13	0.017	0.84
May-21-13	10.6	7.80	281	0.14	2.37	2.28	0.019	0.84
May-28-13	9.1	7.85	269	0.12	2.44	2.45	0.019	0.76
Jun-04-13	9.2	7.65	289	0.24	2.32	2.29	0.017	0.75
Jun-11-13	11.5	7.73	309	0.17	1.88	1.77	0.015	0.84
Jun-18-13	11.7	7.81	302	0.20	2.05	2.07	0.012	0.59
Jun-25-13	10.6	7.53	302	0.13	1.79	1.69	0.012	0.68
Jul-02-13	17.0	7.71	306	0.25	1.95	1.70	0.012	0.70
Jul-09-13	10.8	7.29	310	0.17	1.98	1.97	0.012	0.63
Jul-16-13	13.4	7.52	318	0.28	2.00	2.00	0.013	0.67
Jul-23-13	9.9	7.34	310	0.16	1.83	1.87	0.012	0.62
Jul-30-13	10.4	7.42	313	0.15	2.00	1.73	0.012	0.62
Aug-06-13	12.1	7.57	311	0.14	2.22	2.10	0.011	0.51
Aug-13-13	12.1	7.38	313	0.07	2.02	1.97	0.014	0.68
Aug-20-13	10.4	7.43	316	0.08	2.03	2.02	0.013	0.66
Aug-27-13	16.0	7.46	312	0.09	2.13	2.10	0.017	0.82
Sep-03-13	16.7	7.28	312	0.07	2.18	2.14	0.086	0.39
Sep-10-13	10.0	7.29	308	0.18	2.03	1.93	0.012	0.59
Sep-17-13	14.1	7.30	308	0.08	2.07	2.06	0.084	0.42
Sep-24-13	11.5	7.25	308	0.04	2.12	2.02	0.007	0.33
Oct-01-13	16.7	7.33	310	0.09	2.21	2.25	0.004	0.19
Oct-08-13	17.0	7.69	308	0.09	2.19	2.16	0.003	0.13
Oct-15-13	17.1	7.65	306	0.08	2.19	2.13	0.007	0.32
Oct-22-13	8.2	7.26	316	0.02	1.98	1.92	0.005	0.22

Table D-3. BACC#3 effluent water quality data (con't)

Date	Temperature (°C)	pH	Conductivity (µS/cm)	Turbidity (NTU)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
Oct-29-13	7.0	7.47	317	0.03	1.92	1.90	0.004	0.18
Nov-05-13	8.0	7.51	310	0.03	1.90	1.87	0.003	0.18
Nov-12-13	6.6	7.43	310	0.03	2.15	2.13	0.006	0.26
Nov-19-13	7.6	7.48	314	0.04	1.83	1.85	0.010	0.56
Nov-26-13	6.2	7.63	308	0.02	1.70	1.67	0.037	0.22
Dec-03-13	6.0	7.25	328	0.05	1.64	1.62	0.009	0.56
Dec-10-13	3.4	7.36	324	0.10	1.64	1.63	0.006	0.37
Dec-17-13	3.5	7.17	327	0.72	1.58	1.56	0.009	0.55
Jan-06-14	3.1	7.29	354	0.14	1.66	1.62	0.017	1.06
Jan-14-14	3.3	7.48	329	0.09	1.60	1.58	0.014	0.85
Jan-21-14	3.8	7.18	368	0.09	1.79	1.76	0.010	0.56
Jan-28-14	3.8	7.48	312	0.04	1.63	1.63	0.013	0.78
Feb-04-14	5.2	7.55	319	0.11	1.70	1.65	0.011	0.68
Feb-11-14	4.4	7.28	314	0.05	1.62	1.63	0.015	0.89
Feb-18-14	4.3	7.39	327	0.35	1.58	1.52	0.013	0.85
Feb-25-14	5.1	7.22	315	0.05	1.55	1.55	0.014	0.92
Mar-04-14	5.2	7.47	317	0.04	1.61	1.60	0.014	0.90
Mar-11-14	4.7	7.77	346	0.08	1.67	1.63	0.013	0.76
Mar-18-14	4.6	7.42	330	0.06	1.62	1.55	0.015	0.94
Mar-25-14	5.1	7.38	384	0.08	2.16	2.16	0.014	0.63
Apr-01-14	6.2	7.36	406	0.17	2.16	2.11	0.016	0.77
Apr-08-14	5.8	7.24	351	0.26	1.93	1.82	0.015	0.81
Apr-15-14	7.8	7.22	360	0.12	2.10	2.10	0.009	0.40
Apr-22-14	8.5	7.34	359	0.14	1.74	1.75	0.010	0.57
Apr-29-14	7.9	7.22	344	0.12	1.71	1.69	0.009	0.52
May-07-14	8.7	7.44	370	0.15	1.80	1.76	0.015	0.84
May-13-14	9.8	7.20	345	0.14	1.77	1.71	0.011	0.65
May-20-14	9.7	7.39	341	0.10	1.65	1.63	0.007	0.45

Table D-4. UF#42 permeate water quality data

Date	Temperature (°C)	pH	Conductivity (µS/cm)	Turbidity (NTU)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
Jan-29-13	3.3	7.51	279	0.010	1.90	1.85	0.024	1.26
Feb-12-13	3.1	7.81	262	0.011	1.81	1.73	0.019	0.95
Feb-26-13	4.0	7.67	326	0.014	2.15	2.05	0.027	1.48
Mar-26-13	4.0	7.61	299	0.012	2.01	2.03	0.019	0.96
Apr-09-13	5.4	7.70	295	0.012	2.00	2.00	0.017	0.85
Apr-16-13	6.2	7.64	326	0.012	2.16	2.04	0.028	1.28
May-14-13	7.7	7.68	283	0.012	1.86	1.72	0.017	0.98
May-21-13	9.5	7.73	281	0.012	1.66	1.51	0.017	1.12
May-28-13	9.0	7.78	279	0.012	2.07	2.05	0.016	0.79
Jun-04-13	9.4	7.66	293	0.012	1.92	1.91	0.012	0.61
Jun-11-13	12.1	7.62	309	0.010	1.75	1.63	0.011	0.63
Jun-18-13	12.0	7.73	303	0.012	1.82	1.47	0.011	0.60
Jun-25-13	11.0	7.58	303	0.010	1.45	1.31	0.010	0.77
Jul-02-13	18.0	7.54	306	0.010	1.66	1.66	0.010	0.61
Jul-09-13	11.0	7.51	311	0.010	1.51	1.26	0.013	0.99
Jul-16-13	13.6	7.57	319	0.014	1.80	1.80	0.013	0.99
Jul-23-13	9.8	7.48	312	0.010	1.58	1.58	0.013	0.79
Jul-30-13	10.8	7.49	311	0.010	1.84	1.84	0.012	0.67
Aug-06-13	12.4	7.60	312	0.010	1.85	1.60	0.011	0.66
Aug-13-13	12.5	7.49	311	0.010	1.74	1.73	0.013	0.77
Aug-20-13	10.6	7.42	316	0.011	1.87	1.77	0.013	0.72
Aug-27-13	16.0	7.54	314	0.010	1.83	1.82	0.003	0.18
Sep-03-13	16.4	7.51	314	0.010	1.83	1.83	0.007	0.40
Sep-10-13	11.0	7.40	311	0.010	1.72	1.72	0.012	0.71
Sep-17-13	13.6	7.37	311	0.010	1.80	1.78	0.007	0.38
Sep-24-13	11.7	7.31	311	0.010	1.78	1.78	0.006	0.35
Oct-01-13	17.5	7.42	310	0.010	1.90	1.91	0.002	0.11
Oct-08-13	17.3	7.65	308	0.009	1.88	1.89	0.010	0.50
Oct-15-13	17.6	7.60	308	0.009	1.86	1.86	0.004	0.23
Oct-22-13	8.7	7.43	310	0.010	2.58	1.85	0.003	0.17

Table D-4. UF#42 permeate water quality data (con't)

Date	Temperature (°C)	pH	Conductivity (µS/cm)	Turbidity (NTU)	TOC (mg/L)	DOC (mg/L)	UV$_{254}$ (cm^{-1})	SUVA (L/(mg C·m))
Oct-29-13	8.7	7.44	317	0.010	1.70	1.67	0.008	0.50
Nov-05-13	6.7	7.58	312	0.010	1.88	1.86	0.006	0.37
Nov-12-13	6.7	7.42	307	0.010	2.10	1.97	0.004	0.18
Nov-19-13	6.6	7.43	311	0.010	1.68	1.75	0.010	0.58
Nov-26-13	6.0	7.52	312	0.010	1.51	1.52	0.006	0.41
Dec-03-13	6.2	7.34	326	0.010	1.44	1.44	0.009	0.63
Dec-10-13	5.0	7.43	326	0.010	1.44	1.41	0.006	0.43
Dec-17-13	3.9	7.28	325	0.010	1.42	1.40	0.009	0.60
Jan-06-14	3.5	7.39	358	0.010	1.43	1.44	0.016	1.13
Jan-14-14	3.5	7.51	331	0.010	1.48	1.41	0.015	1.06
Jan-21-14	3.8	7.25	371	0.010	1.75	1.77	0.010	0.58
Jan-28-14	4.1	7.73	309	0.010	1.48	1.46	0.013	0.86
Feb-04-14	5.5	7.79	315	0.011	1.50	1.47	0.016	1.07
Feb-11-14	4.9	7.46	312	0.011	1.55	1.42	0.015	1.04
Feb-18-14	4.4	7.47	329	0.010	1.43	1.41	0.013	0.95
Feb-25-14	5.3	7.28	313	0.010	1.43	1.41	0.015	1.09
Mar-04-14	5.0	7.45	320	0.010	1.46	1.46	0.013	0.91
Mar-11-14	4.9	7.74	344	0.010	1.44	1.45	0.013	0.87
Mar-18-14	4.4	7.47	333	0.010	2.02	1.94	0.016	0.78
Mar-25-14	5.0	7.47	390	0.010	1.79	1.76	0.014	0.80
Apr-01-14	6.2	7.40	405	0.010	1.81	1.73	0.016	0.92
Apr-08-14	6.0	7.37	355	0.010	1.67	1.60	0.015	0.95
Apr-15-14	7.6	7.56	360	0.010	1.78	1.78	0.010	0.58
Apr-22-14	8.3	7.34	359	0.010	1.57	1.56	0.011	0.69
Apr-29-14	8.0	7.28	345	0.010	1.52	1.52	0.010	0.63
May-07-14	8.7	7.40	372	0.010	1.61	1.58	0.015	0.97
May-13-14	9.9	7.29	350	0.010	1.88	1.62	0.011	0.65
May-20-14	10.0	7.40	342	0.010	1.48	1.48	0.007	0.50

Table D-5. UF Pilot#1 permeate water quality data

Date	Temperature (°C)	pH	Conductivity (µS/cm)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
May-21-13	9.6	7.74	282	2.10	2.05	0.017	0.67
May-28-13	9.0	7.89	269	2.04	2.06	0.019	0.72
Jun-04-13	9.4	7.60	293	1.92	1.84	0.012	0.65
Jun-11-13	12.1	7.58	311	1.84	1.74	0.011	0.61
Jun-18-13	12.0	7.65	306	2.43	2.30	0.016	0.68
Jun-25-13	11.1	7.57	303	1.38	1.39	0.011	0.79
Jul-02-13	17.1	7.50	308	1.66	1.66	0.023	0.28
Jul-09-13	10.6	7.47	313	1.71	1.50	0.011	0.73
Jul-16-13	13.2	7.58	318	1.80	1.80	0.013	0.77
Jul-23-13	10.7	7.46	312	1.67	1.54	0.011	0.60
Jul-30-13	11.3	7.48	309	1.86	1.78	0.012	0.64
Aug-06-13	12.0	7.53	314	1.92	1.89	0.014	0.77
Aug-13-13	12.4	7.47	312	1.76	1.75	0.014	0.77
Aug-20-13	10.5	7.48	317	1.79	1.72	0.013	0.78
Aug-27-13	16.1	7.56	313	1.86	1.83	0.003	0.15
Sep-03-13	16.7	7.41	312	1.86	1.84	0.006	0.30
Sep-10-13	10.6	7.41	309	2.20	2.19	0.013	0.59
Sep-17-13	12.6	7.37	309	2.33	2.33	0.007	0.30
Sep-24-13	13.2	7.35	309	1.84	1.83	0.005	0.26
Oct-01-13	13.2	7.47	310	1.94	1.96	0.003	0.15
Oct-08-13	18.0	7.55	308	1.86	1.86	0.011	0.59
Oct-15-13	17.3	7.51	308	2.00	1.94	0.004	0.19
Oct-22-13	8.4	7.43	316	1.91	1.87	0.002	0.11
Oct-29-13	8.9	7.52	366	1.68	1.67	0.007	0.41
Nov-05-13	6.7	7.58	312	1.70	1.68	0.006	0.37
Nov-12-13	6.7	7.48	334	1.93	1.93	0.013	0.66
Nov-19-13	7.7	7.45	314	1.73	1.72	0.010	0.57
Nov-26-13	6.4	7.47	316	1.54	1.49	0.008	0.51

Table D-5. UF Pilot#1 permeate water quality data (con’t)

Date	Temperature (°C)	pH	Conductivity (µS/cm)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
Dec-03-13	6.4	7.48	353	1.39	1.38	0.014	1.01
Dec-10-13	5.9	7.48	340	1.38	1.36	0.057	0.51
Dec-17-13	3.3	7.30	327	1.39	1.39	0.009	0.67
Jan-06-14	2.5	7.36	350	1.49	1.44	0.017	1.17
Jan-14-14	3.0	7.48	340	1.43	1.42	0.015	1.02
Jan-21-14	3.8	7.24	361	1.80	1.81	0.010	0.58
Jan-28-14	4.2	7.63	313	1.54	1.55	0.013	0.82
Feb-04-14	5.2	7.71	324	1.42	1.39	0.012	0.86
Feb-11-14	4.9	7.39	311	1.50	1.44	0.015	1.04
Feb-18-14	4.3	7.47	327	1.45	1.41	0.013	0.92
Feb-25-14	4.8	7.26	313	1.37	1.36	0.015	1.07
Mar-04-14	4.8	7.45	321	1.48	1.43	0.014	0.95
Mar-11-14	5.2	7.71	342	1.40	1.40	0.012	0.86
Mar-18-14	5.7	7.53	333	1.76	1.53	0.016	1.07
Mar-25-14	5.2	7.44	385	1.78	1.67	0.014	0.86
Apr-01-14	6.2	7.36	405	1.80	1.76	0.015	0.87
Apr-08-14	5.9	7.37	355	1.80	1.76	0.019	1.07
Apr-15-14	7.6	7.46	363	1.88	1.78	0.010	0.51
Apr-22-14	8.2	7.37	360	1.58	1.57	0.011	0.72
Apr-29-14	8.6	7.39	344	1.51	1.50	0.009	0.59
May-07-14	8.7	7.41	371	1.56	1.54	0.015	0.95
May-13-14	10.4	7.30	351	1.51	1.52	0.011	0.69
May-20-14	10.1	7.40	343	1.47	1.48	0.008	0.55

Table D-6. UF Pilot#2 permeate water quality data

Date	Temperature (°C)	pH	Conductivity (µS/cm)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
Mar-11-14	7.7	7.91	355	2.33	2.32	0.018	0.76
Mar-18-14	6.0	7.54	336	1.67	1.68	0.017	0.98
Mar-25-14	6.2	7.90	426	1.90	1.81	0.027	1.44
Apr-01-14	7.7	7.49	419	1.96	1.97	0.015	0.78
Apr-08-14	8.2	7.54	371	1.79	1.78	0.019	1.04
Apr-15-14	9.7	7.98	453	2.17	2.00	0.037	1.72
Apr-22-14	9.9	7.42	355	1.66	1.67	0.018	1.07
Apr-29-14	10.6	7.36	355	1.51	1.51	0.017	1.15
May-07-14	8.7	7.58	391	1.74	1.70	0.024	1.43
May-13-14	12.0	7.32	354	1.63	1.62	0.019	1.20
May-20-14	11.4	7.64	363	1.55	1.45	0.017	1.12

Table D-7. UF Pilot#3 permeate water quality data

Date	Temperature (°C)	pH	Conductivity (µS/cm)	TOC (mg/L)	DOC (mg/L)	UV_{254} (cm^{-1})	SUVA (L/(mg C·m))
Apr-01-14	5.2	7.44	382	1.79	1.78	0.014	0.78
Apr-08-14	5.8	7.44	353	1.84	1.78	0.015	0.84
Apr-15-14	9.7	7.57	363	1.95	1.92	0.010	0.51
Apr-22-14	8.4	7.35	361	1.57	1.54	0.011	0.74
Apr-29-14	8.2	7.37	342	1.56	1.56	0.011	0.67
May-07-14	10.5	7.42	370	1.61	1.56	0.015	0.96
May-13-14	10.6	7.32	351	1.52	1.52	0.011	0.69
May-20-14	10.3	7.57	339	1.41	1.40	0.007	0.51

Appendix E: LC-OCD Raw Data

Table E-1. LC-OCD biopolymer data

Date	Biopolymer concentration (μg/L)					
	Raw water	Ozone effluent	BACC#3 effluent	UF#42 permeate	UF Pilot#1 permeate	UF Pilot#2 permeate
Jan-29-13	207	213	237	67	-	-
Feb-12-13	257	229	268	101	-	-
Feb-26-13	291	269	260	55	-	-
Mar-26-13	268	238	129	70	-	-
Apr-09-13	252	240	234	76	-	-
Apr-16-13	300	288	299	125	-	-
May-14-13	215	216	212	63	-	-
May-21-13	260	259	269	94	76	-
May-28-13	253	255	260	118	62	-
Jun-04-13	245	223	235	90	72	-
Jun-11-13	247	238	286	138	147	-
Jun-18-13	276	259	259	110	108	-
Jun-25-13	259	268	265	113	138	-
Jul-02-13	337	324	318	158	149	-
Jul-09-13	266	260	273	103	106	-
Jul-16-13	288	282	286	103	109	-
Jul-23-13	272	263	256	88	180	-
Jul-30-13	290	289	266	129	134	-
Aug-06-13	342	332	308	102	131	-
Aug-13-13	354	349	354	141	155	-
Aug-20-13	306	284	290	108	115	-
Aug-27-13	358	351	351	141	155	-
Sep-03-13	335	353	337	139	148	-
Sep-10-13	258	266	261	107	122	-
Sep-17-13	314	322	293	110	115	-
Sep-24-13	267	266	269	93	107	-
Oct-01-13	363	397	401	162	184	-
Oct-08-13	378	341	369	135	139	-

Table E-1. LC-OCD biopolymer data (con’t)

Date	Biopolymer concentration (µg/L)					
	Raw water	Ozone effluent	BACC#3 effluent	UF#42 permeate	UF Pilot#1 permeate	UF Pilot#2 permeate
Oct-15-13	368	357	355	142	169	-
Oct-22-13	228	225	229	92	101	-
Oct-29-13	226	225	226	89	116	-
Nov-05-13	224	228	212	62	100	-
Nov-12-13	239	240	213	98	112	-
Nov-19-13	238	229	238	88	101	-
Nov-26-13	219	217	215	74	94	-
Dec-03-13	299	290	282	117	107	-
Dec-10-13	288	256	269	124	128	-
Dec-17-13	288	280	288	99	90	-
Jan-06-14	283	283	268	73	183	-
Jan-14-14	256	241	258	79	102	-
Jan-21-14	258	266	253	62	86	-
Jan-28-14	272	262	277	75	84	-
Feb-04-14	236	260	326	75	108	-
Feb-11-14	280	283	286	112	99	-
Feb-18-14	255	261	273	84	90	-
Feb-25-14	232	236	236	100	86	-
Mar-04-14	279	252	273	92	124	-
Mar-11-14	218	214	273	77	82	109
Mar-18-14	236	236	232	68	99	126
Mar-25-14	295	244	224	72	88	167
Apr-01-14	293	311	322	129	142	201
Apr-08-14	225	239	268	100	95	201
Apr-15-14	249	227	243	86	81	210
Apr-22-14	290	259	255	85	98	145
Apr-29-14	241	181	180	66	124	103
May-07-14	307	275	283	91	93	189
May-13-14	289	269	272	81	82	63
May-20-14	329	290	300	96	103	92

Table E-2. LC-OCD humic substances data

Date	Humic substances concentration (µg/L)					
	Raw water	Ozone effluent	BACC#3 effluent	UF#42 permeate	UF Pilot#1 permeate	UF Pilot#2 permeate
Jan-29-13	1074	1062	1108	1174	-	-
Feb-12-13	1013	1010	1049	1040	-	-
Feb-26-13	1012	1030	1015	845	-	-
Mar-26-13	1159	1006	1424	1489	-	-
Apr-09-13	986	1007	1019	1006	-	-
Apr-16-13	1096	1054	1267	1095	-	-
May-14-13	982	1001	1004	991	-	-
May-21-13	1050	1067	1096	1045	1115	-
May-28-13	1220	991	1018	1229	998	-
Jun-04-13	1051	1060	1035	873	1032	-
Jun-11-13	1137	1053	1278	948	1020	-
Jun-18-13	1006	979	951	952	1021	-
Jun-25-13	1018	984	951	980	979	-
Jul-02-13	1102	1044	1021	1023	1045	-
Jul-09-13	955	1063	955	947	915	-
Jul-16-13	1094	1074	1037	1018	994	-
Jul-23-13	1011	1008	928	941	963	-
Jul-30-13	991	1024	917	929	982	-
Aug-06-13	1026	915	1073	1037	905	-
Aug-13-13	1034	1000	926	952	977	-
Aug-20-13	1069	1023	866	885	896	-
Aug-27-13	1049	981	955	994	1008	-
Sep-03-13	1173	1114	1083	1067	1068	-
Sep-10-13	1017	959	912	958	1013	-
Sep-17-13	958	1071	1063	1087	958	-
Sep-24-13	992	935	893	918	911	-
Oct-01-13	1562	1390	1381	1028	1121	-
Oct-08-13	990	1167	955	969	997	-
Oct-15-13	1175	1110	1111	1086	996	-
Oct-22-13	933	1107	1039	942	899	-

Table E-2. LC-OCD humic substances data (con’t)

Date	Humic substances concentration (μg/L)					
	Raw water	Ozone effluent	BACC#3 effluent	UF#42 permeate	UF Pilot#1 permeate	UF Pilot#2 permeate
Oct-29-13	1150	986	936	949	997	-
Nov-05-13	954	1054	1042	902	990	-
Nov-12-13	1080	1014	1008	1025	1020	-
Nov-19-13	965	995	922	981	912	-
Nov-26-13	1006	1150	923	944	879	-
Dec-03-13	1118	1164	1088	1017	1029	-
Dec-10-13	1102	1133	977	974	930	-
Dec-17-13	1042	970	927	960	918	-
Jan-06-14	1132	1131	907	917	1823	-
Jan-14-14	1062	1027	1002	978	1031	-
Jan-21-14	1036	1036	1020	1055	1026	-
Jan-28-14	1022	1018	1048	1046	1205	-
Feb-04-14	1043	1026	1091	1015	1019	-
Feb-11-14	982	990	965	988	984	-
Feb-18-14	1002	1009	997	999	1024	-
Feb-25-14	1002	1005	1013	1017	1011	-
Mar-04-14	994	1075	1002	986	998	-
Mar-11-14	1009	1147	972	1046	1032	1083
Mar-18-14	982	930	934	957	975	988
Mar-25-14	1048	1047	1019	1026	1122	1122
Apr-01-14	1247	1004	996	1049	1029	1092
Apr-08-14	1032	1002	1010	962	1020	1089
Apr-15-14	1048	1083	1081	1087	1082	1230
Apr-22-14	1016	1107	1087	1134	1015	1037
Apr-29-14	976	958	946	958	963	986
May-07-14	1070	1050	1049	1058	1058	1101
May-13-14	1076	1048	1029	1033	1033	1097
May-20-14	984	948	934	1066	1086	948

Table E-3. LC-OCD building blocks data

Date	Building blocks concentration (μg/L)					
	Raw water	Ozone effluent	BACC#3 effluent	UF#42 permeate	UF Pilot#1 permeate	UF Pilot#2 permeate
Jan-29-13	357	363	346	273	-	-
Feb-12-13	406	418	400	390	-	-
Feb-26-13	473	467	463	460	-	-
Mar-26-13	276	401	121	77	-	-
Apr-09-13	415	372	382	368	-	-
Apr-16-13	486	510	311	535	-	-
May-14-13	392	385	370	393	-	-
May-21-13	391	412	327	374	211	-
May-28-13	269	425	428	436	404	-
Jun-04-13	293	388	550	327	430	-
Jun-11-13	328	353	177	377	228	-
Jun-18-13	198	298	199	181	339	-
Jun-25-13	396	363	375	299	298	-
Jul-02-13	387	397	387	363	402	-
Jul-09-13	430	271	372	354	375	-
Jul-16-13	411	358	379	372	407	-
Jul-23-13	403	362	366	391	263	-
Jul-30-13	405	403	364	384	339	-
Aug-06-13	388	418	320	303	439	-
Aug-13-13	369	397	428	360	356	-
Aug-20-13	341	363	465	431	442	-
Aug-27-13	414	424	406	401	414	-
Sep-03-13	306	344	323	286	297	-
Sep-10-13	388	410	354	354	506	-
Sep-17-13	509	365	345	290	552	-
Sep-24-13	463	438	431	412	406	-
Oct-01-13	191	72	51	398	289	-
Oct-08-13	486	279	416	423	381	-
Oct-15-13	318	346	236	288	401	-
Oct-22-13	446	259	255	356	434	-

Table E-3. LC-OCD building blocks data (con’t)

Date	Building blocks concentration (μg/L)					
	Raw water	Ozone effluent	BACC#3 effluent	UF#42 permeate	UF Pilot#1 permeate	UF Pilot#2 permeate
Oct-29-13	238	379	329	322	383	-
Nov-05-13	430	271	215	380	300	-
Nov-12-13	452	485	435	387	300	-
Nov-19-13	398	362	365	396	367	-
Nov-26-13	377	190	376	343	390	-
Dec-03-13	350	281	310	345	394	-
Dec-10-13	333	358	516	434	421	-
Dec-17-13	419	476	457	463	483	-
Jan-06-14	361	327	565	543	383	-
Jan-14-14	380	448	412	431	362	-
Jan-21-14	462	472	470	452	473	-
Jan-28-14	395	396	325	383	229	-
Feb-04-14	365	380	241	423	353	-
Feb-11-14	423	417	411	401	408	-
Feb-18-14	464	456	488	475	432	-
Feb-25-14	436	427	454	444	440	-
Mar-04-14	384	425	359	409	374	-
Mar-11-14	389	245	428	357	340	401
Mar-18-14	385	422	424	421	358	366
Mar-25-14	399	415	379	402	326	515
Apr-01-14	252	473	466	411	401	449
Apr-08-14	395	427	355	432	389	422
Apr-15-14	400	400	441	423	374	548
Apr-22-14	469	365	417	442	424	439
Apr-29-14	500	526	488	418	377	420
May-07-14	445	455	426	426	413	479
May-13-14	391	411	392	406	391	388
May-20-14	463	477	450	328	326	522

Table E-4. LC-OCD LMW neutrals data

Date	LMW neutrals concentration (µg/L)					
	Raw water	Ozone effluent	BACC#3 effluent	UF#42 permeate	UF Pilot#1 permeate	UF Pilot#2 permeate
Jan-29-13	208	199	213	177	-	-
Feb-12-13	232	169	205	194	-	-
Feb-26-13	224	261	201	172	-	-
Mar-26-13	217	208	215	199	-	-
Apr-09-13	204	191	192	261	-	-
Apr-16-13	244	247	246	229	-	-
May-14-13	177	175	182	147	-	-
May-21-13	176	190	202	206	111	-
May-28-13	242	193	206	264	155	-
Jun-04-13	203	189	206	150	184	-
Jun-11-13	214	225	210	196	224	-
Jun-18-13	213	218	230	241	207	-
Jun-25-13	213	218	230	241	207	-
Jul-02-13	219	218	210	128	254	-
Jul-09-13	190	171	199	165	151	-
Jul-16-13	246	227	211	147	189	-
Jul-23-13	208	223	165	159	159	-
Jul-30-13	187	270	214	156	163	-
Aug-06-13	177	200	171	139	153	-
Aug-13-13	208	213	184	142	155	-
Aug-20-13	305	183	194	147	148	-
Aug-27-13	255	230	194	157	159	-
Sep-03-13	197	215	184	146	149	-
Sep-10-13	171	184	173	146	242	-
Sep-17-13	186	177	191	168	328	-
Sep-24-13	333	184	170	161	146	-
Oct-01-13	210	184	187	155	208	-
Oct-08-13	188	156	185	155	155	-
Oct-15-13	180	251	182	156	152	-
Oct-22-13	173	164	143	149	160	-

Table E-4. LC-OCD LMW neutrals data (con’t)

Date	LMW neutrals concentration (µg/L)					
	Raw water	Ozone effluent	BACC#3 effluent	UF#42 permeate	UF Pilot#1 permeate	UF Pilot#2 permeate
Oct-29-13	181	155	243	137	181	-
Nov-05-13	216	172	153	129	150	-
Nov-12-13	197	206	179	156	160	-
Nov-19-13	173	183	169	403	144	-
Nov-26-13	200	191	182	154	142	-
Dec-03-13	248	219	245	176	282	-
Dec-10-13	231	249	264	203	184	-
Dec-17-13	184	189	193	176	165	-
Jan-06-14	226	205	206	205	358	-
Jan-14-14	187	225	182	164	150	-
Jan-21-14	199	193	190	169	190	-
Jan-28-14	194	203	192	170	154	-
Feb-04-14	231	216	182	178	172	-
Feb-11-14	183	184	208	174	239	-
Feb-18-14	194	200	191	176	172	-
Feb-25-14	204	188	199	178	167	-
Mar-04-14	215	202	149	139	156	-
Mar-11-14	253	341	341	188	154	136
Mar-18-14	187	192	190	174	264	447
Mar-25-14	361	343	134	191	174	568
Apr-01-14	224	204	224	177	193	308
Apr-08-14	206	211	201	185	191	352
Apr-15-14	220	235	237	207	196	354
Apr-22-14	239	223	290	202	200	217
Apr-29-14	223	228	198	135	177	234
May-07-14	211	199	204	229	206	401
May-13-14	214	213	209	187	190	181
May-20-14	166	204	191	176	172	513

LMW- Low molecular weight

Table E-5. UF Pilot#3 permeate LC-OCD data

Date	Concentration (µg/L)			
	Biopolymers	Humic substances	Building blocks	LMW neutrals
Apr-15-2014	208	1121	431	420
Apr-22-2014	127	1002	439	198
Apr-29-2014	128	983	402	167
May-07-2014	84	1034	423	194
May-13-2014	101	1045	391	200
May-20-2014	90	1063	310	164

Table E-6. UF Pilot#1 and Pilot#2 backpulse sample LC-OCD data

Date	Concentration (µg/L)				
	UF Pilot	Biopolymers	Humic substances	Building blocks	LMW neutrals
May-13-2014	#1	274	993	369	224
May-13-2014	#1	287	1060	398	250
May-13-2014	#2	508	1137	414	244
May-13-2014	#2	607	1146	423	225
May-20-2014	#1	405	928	463	151
May-20-2014	#1	330	853	453	170
May-20-2014	#2	356	936	492	521
May-20-2014	#2	377	913	507	548

Table E-7. Raw water at intake screen pre-chlorination LC-OCD data

Date	Concentration (µg/L)			
	Biopolymers	Humic substances	Building blocks	LMW neutrals
Feb-04-2014	260	1019	401	224
Feb-11-2014	294	976	421	209
Feb-18-2014	249	1023	477	193
Feb-25-2014	250	1028	472	221
Mar-04-2014	273	974	417	194
Mar-18-2014	235	949	416	191